Quantification of the methane emission from three UK landfills

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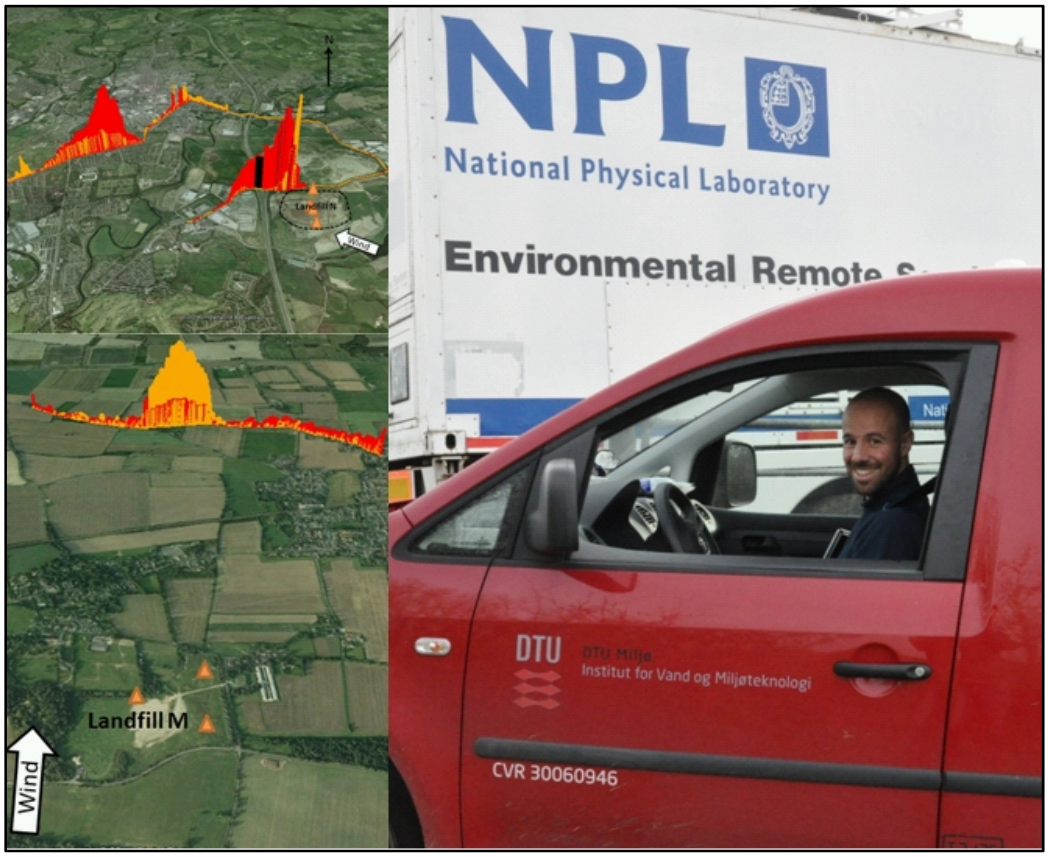

Left: The relative atmospheric concentration of methane and tracer gas downwind from two UK landfills. Right: the two vehicles from DTU Environment and NPL performing methane measurements in parallel.

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Contents

1. Introduction and purpose

The Technical University of Denmark (DTU) has recently implemented a novel analytical setup enabling mobile measurements of small changes (ppb level) in atmospheric methane and acetylene concentrations. This enables detection and quantification of methane sources by performing measurements downwind from the source in combination with release and measurement of acetylene as a tracer gas. The analytical setup and the dynamic tracer dispersion method have been tested at approximately 20 Danish landfills since November 2011 (Mønster et al., 2014a; Mønster et al., 2014b), building up a sound knowledge on quantification of the total fugitive methane emission from landfills. Danish landfills are, on an average, relatively small compared to UK landfills, and two measurement campaigns performed during summer 2014 at an UK landfill led to an agreement with DEFRA to perform methane quantification measurements on three UK landfills, where two of the measurement campaigns were done in parallel with the National Physics Laboratory (NPL), who were using the DiAL methodology. This technical report presents the measured methane emissions from the three landfills, and is a part of a joined report submitted by NPL.

The objective of this study was to quantify the total methane emission from three UK landfills and to compare these emission rates with the emissions rates obtained by DiAL measurements carried out by NPL. The measurement campaigns were conducted from November 10th to November 21st and included more than 100 successful methane plume traverses.

2. Description of the landfills

The three landfills are anonymous in this report and the descriptions of them are therefore not detailed. The landfills are named M, J and N and the following is a short description of the individual landfills.

Landfill M is placed in an old sand and gravel quarry. The landfill area is approximately 15 ha where approximately 1,000,000 m^3 domestic, commercial and low level radioactive wastes was deposited between 1972 and 1988 where after the landfill was closed. The landfill has no liner and the top is covered with topsoil and pulverised waste. The waste thickness varies between 10 and 20 meters. Landfill gas is extracted 8 hours per day and flared. The flaring rate was approximately 25 kg methane h^{-1} during the days where the measurements were performed. The landfill area is currently open to the public and is frequently used for dog walking by the local residences.

The operation of landfill J was started in the early 1970's. Initially, the landfill only accepted inert material, but in 1989 the site started also to accept commercial and industrial waste. Landfill J is still active and continuously receiving waste - no information is available on the individual waste cells with regard to disposal start and finish dates. The total volume of deposited waste up to present time is approximately 16 million m^3. The landfill has gas recovery and utilization in onsite gas engines (a new and an older series of engines, both still burning landfill gas for electricity production). The amount of methane recovered was around 86 kg h^{-1}. The landfill also has onsite leachate treatment.

Landfill N is an active landfill placed in an old quarry, which was started in 2005. The landfill is situated next to an older closed landfill (1989 to 2005), and both landfills have gas recovery, which utilized gas engines for power generation. On the measurement day (November 21st), a landfill gas flow of 2683 m^3 h^{-1} (normalised to 50% methane) was recovered, which is approximately 956 kg methane h^{-1}. The recovered methane comes from both the old and the new landfill and is mixed and utilized together. The old, covered landfill contains about 7.5 Tg waste, mainly consisting commercial and industrial wastes. The new landfill (landfill N) has a total capacity of 7.8 million m^3, which should allow waste depositing until 2026-27. It has currently about 3.8 Tg waste, predominantly non-hazardous commercial and industrial wastes and domestic wastes.

3. Dynamic plume measurement using mobile analytical platforms

Total landfill methane emissions were quantified using a mobile tracer dispersion method that combines a controlled release of tracer gas from the landfill with concentration measurements downwind of the landfill, by using a mobile high-resolution analytical instrument (Börjesson et al., 2007, 2009; Galle et al., 2001; Scheutz et al., 2011). The method has been used successfully in the last few decades, and with new developments in analytical technology it has become a powerful tool for quantifying methane emissions from landfills (Mønster et al., 2014; 2015). The tracer dispersion method in general is based on the assumption that a tracer gas released at an emission source, in this case a landfill, will disperse in the atmosphere in the same way as methane emitted from the landfill. Assuming a defined wind direction, well mixed air above the landfill for the methane and tracer gas to be fully mixed, and a constant tracer gas release, the methane emission rate can be calculated as a function of the ratio of the integrated cross-plume concentration of the emitted methane and the integrated cross-plume concentration of the released tracer gas, as follows:

$$E_{gas} = Q_{tracer} \cdot \frac{\int_{Plume\ end\ 1}^{Plume\ end\ 2} C_{gas}\, dx}{\int_{Plume\ end\ 1}^{Plume\ end\ 2} C_{tracer}\, dx} \cdot \frac{MW_{gas}}{MW_{tracer}} \qquad \text{(Eq. 1)}$$

Where E_{gas} is the methane emission rate (kg h^{-1}), Q_{tracer} is the release rate of the tracer gas (kg h^{-1}), C_{gas} and C_{tracer} denote cross-plume concentrations (ppbv) above the background concentration, MW denotes molecular weights and x corresponds to distance across the plume. The principle is shown in Figure 1.

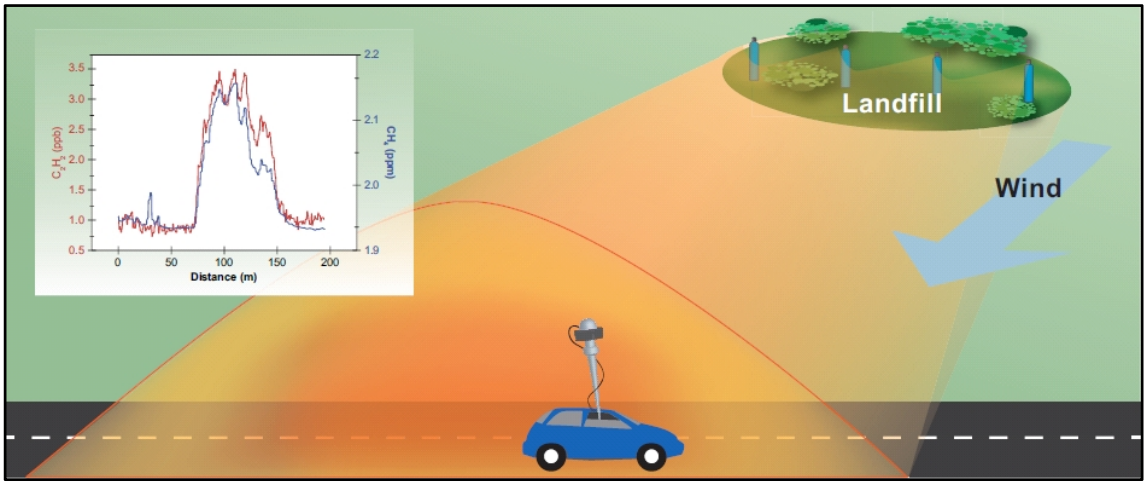

Figure 1. The principle of the dynamic plume method for quantifying greenhouse gas emissions from fugitive sources.

In the current measurement campaigns, the downwind measurements were done at suitable roads near the three landfills, and distances varied from day to day, depending on the wind direction, the degree of dispersion and the accessibility of roads and possible interference with other methane sources in the area. The optimal distance for measuring a site's total emissions depends on the size of the emission area, the topography of the site and weather conditions such as wind speed and sun, leading to a higher dispersion of the methane plume (Mønster et al., 2014). All quantifications at the three landfills were made within a distance of 800 to 5800 m. Quantifications were made by performing multiple transects across the plume and then calculating the methane/tracer ratio (Eq. 1) for each transect. In this way, a change in dilution due to a change in wind speed, or turbulence changing vertical mixing, would be the same for both gasses at each individual plume measurement. At each transect it was ensured that the whole plume was measured before turning the vehicle to measure the plume again. This enabled the establishment of a baseline of background concentrations to be subtracted from the measurements, in order to obtain the landfill's contribution to the plume. Each transect measurement took between 1 and 5 minutes to perform. The transect time depended on the width of the plume at the measurement distance (depending on dispersion and distance) and driving speed. The driving speed was typically 20-30 km h^{-1}, depending on road and traffic conditions.

Measurements were performed with a cavity ring-down spectroscopy (CRDS), methane/acetylene analyser (G2203, Picarro Inc., USA), where acetylene was used as tracer gas. Atmospheric air was sampled from the roof of a vehicle and brought to the analyser via an external pump, thus enabling a fast response time when a plume was entered while driving. The atmospheric concentrations of methane, acetylene and water were measured with a frequency of 2 Hz and logged together with the atmospheric conditions and GPS position. The precision of methane and acetylene concentrations were 0.48 ppb and 0.40 ppb, respectively, making it possible to detect small changes in atmospheric concentrations while driving and monitoring the concentration on the screen attached to the analyser. The CRDS instrument was calibrated by the manufacture. The methane measurements were calibrated using four gravimetrically prepared mixtures of methane with concentration ranging from 1.7 to 2.3 ppm. Ultrapure nitrogen was used as a zero calibration. The acetylene measurement was not calibrated directly with a standard. Instead, the spectroscopy calibration constant that was measured on another acetylene instrument that uses the same spectral line (Model G1203, SN: DFADS002, Picarro, Inc., Santa Clara) was applied to our measurements. That other instrument was calibrated against a 103 ppb acetylene in air mixture

(Linde Premium Products (formerly SpectraGases), Stewartsville, NJ) that was guaranteed with an accuracy of 10%. A series of laboratory tests were then performed to establish the basic performance of the analyzer, consisting of continuous measurements on prepared gas mixture (see Mønster et al11). The CRDS instrument is occasionally tuned by adjusting the focus of the lasers wavelengths to reduce the signal to noise ratio. For more information on the CRDS, see Mønster et al., (2014).

An anemometer (All-In-One weather sensor, model 102780, Climatronics, USA.) was mounted on top of the vehicle, in order to log wind speed and direction, temperature and atmospheric pressure, and a GPS (model R330 GNSS Receiver and A101 Smart Antenna, Hemiphere, Canada) was attached to the front window, in order to log the position of the vehicle measured the location within 20 cm precision.

Gas bottles of 15 L filled with 2.7 kg acetylene were used to release the tracer gas at the landfills, and the flow was controlled with calibrated flow meters (Sho-rate, Brooks Instrument, Holland). The qualitative measurements were done on and around the landfill (when possible) and on all possible roads nearby. The tracer bottles were placed in those areas estimated from the screening measurements to have the highest methane emissions, in order to simulate the emission in the best possible way. Quantification measurements were then done downwind at an appropriate distance from the landfill, far enough to enable a mixing of the tracer gas and methane (good correlation between tracer gas and methane) and close enough to get a good signal-to-noise ratio. More information on the instrumentation, method, influence of incorrect trace gas placement and the distance to the landfill can be found in Mønster et al. (2014).

The uncertainty of the ‘‘true emission rate’’ is a combination of the individual uncertainties, which can be divided into the analytical uncertainty, uncertainty in tracer gas release, data treatment uncertainty and tracer, source and transect geometry errors (Mønster et al., 2014). The overall analytical uncertainty was estimated to be less than 10% driven by the measurement blend uncertainty of C_2H_2. The uncertainty of the tracer gas release was 5%, determined by the uncertainty of the flowmeters. The uncertainty in transect analysis is highly depending on the stability of the background concentration and the signal-to-noise ratio. Our measurements have shown that the tracer dispersion method has reproducibility uncertainty of 1–2% under normal conditions and emissions above 50 kg CH_4 h^{-1} at a 1.2 km distance from the source (Mønster et al., 2014). At lower signal-to-noise ratios, an error in the area due to an erroneous determination of the baseline can become significant. The baseline uncertainty is not necessarily the raw noise of the data, but can be less if the baseline is stable and an average of multiple baseline points can be used to determine the baseline. Changes of background concentration during a measurement can lead to a larger uncertainty on the baseline.

4. Description of the measurement campaigns

Measurements were performed from November 10th to November 21th, 2014 with no measurements in the weekend 14th to 16th of November. Table 1 provides an overview of the performed measurements and the weather conditions. On November 10th to 13th, the methane emission rate was quantified at the first landfill, called landfill M. The weather was windy with a bit of rain and all in all good conditions for quantification with tracer dispersion method. Landfill

J was measured from November 17th to 20th. The weather had cleared up and there was little to no, and unstable wind. These are not optimal conditions for application of the tracer dispersion method. On November 21st, the emission from landfill N was measured. The weather was clouded and rain was approaching, so measurements were done as fast as possible, as the landfill wound be impossible to move around on after a rain fall.

Table 1. Overview of the performed measurements and the conditions during the measurement periods. Weather data is from official nearby weather stations accessed through www.wunderground.com.

Date	Time	Landfill	wind speed ($m\ s^{-1}$) and direction	Average temperature (°C)	Average pressure (hPa)	Tracer gas release (bottles/total release in $kg\ h^{-1}$)	Plume traverses	Measurement distance (m)	Gas collection
Nov. 11th	18:00 – 19:15	M	3.5 SSE	12.3	997.9	3/2.67	9	800 & 1600	Off (on 8:00 to 16:00)
Nov. 12th	12:30 – 14.30		4.2 S	14.2	998.2	3/2.67	29	1500 to 2000	Flaring estimated: 23 $kg\ h^{-1}$
Nov. 13th	11:00 – 11:30 & 16:45 – 18:15		4.1 & 3.9 SSE	13.1 & 12.1	1006.7 & 1006.0	3/2.68	10 & 10	1500 to 1800	Off (on 8:00 to 10:10)
Nov. 17th	16:30 – 17:30	J	1.8 E	8.7	999.2	3/2.68	7	5600	Gas recovery 2064 $kg\ h^{-1}$
Nov. 18th	16:30 – 17:30		2.9 ENE	9.6	1009.4	2/1.82	7	5800	Gas recovery 2075 $kg\ h^{-1}$
Nov. 19th	13:30 – 15:00		1.7 SE	13.0	1017.2	2/1.82	7	2500 & 3900	Gas recovery 2031 $kg\ h^{-1}$
Nov. 20th	14:15 – 15:30		1.8 E	9.9	1021.9	3/2.79	11	5600	Gas recovery 2106 $kg\ h^{-1}$
Nov. 21st	13:50 – 15:10	N	3.1 ESE	7.9	1018.3	4/3.71	12	2000 to 2500	Gas recovery 956 $kg\ h^{-1}$

5. Description of the data processing.

It was sought to obtain the highest possible number of successful plume traverses at each measurement day at all three landfills measured. Initially, a visual screening was done on all measured plumes to check for interfering methane sources. In very few of the plume traverses, a small methane source right next to the measurement road resulted in a narrow scarp additional increase in the methane concentration, which normally could be removed in the data processing allowing to also use these plumes to calculate the whole landfill emission.

The release of tracer gas was controlled and showed a stable release. At measurements conducted late in the afternoon/evening, the tracer release was left on the landfill and the bottles were allowed to empty completely. To assure that plume measurements were not done while the tracer release was decreasing due to almost empty bottles, conservative time windows for measurements were made.

The plumes passing the visual screening were integrated individually to find the methane/tracer gas ratio for each plume transect, as this has been found to be the most accurate method to obtain

the methane/tracer gas ratio (Mønster et al., 2014). This was also done in cases where the methane and tracer gas plumes did not completely overlap. The ratio of the areas can be used, as measurements were done far from the landfill and both gasses (methane and acetylene) undergo the same atmospheric dispersion.

The emission rate from the individual days was calculated by taking the average of all the emission rates calculated from the accepted plumes rations. The uncertainty of the averaged emission rate was then calculated as the standard error of the mean value on a 95% confidence interval.

6. Results & Discussion

6.1 Landfill M

6.1.1 Methane screening of the area around landfill M

Initial methane screening of the area around the landfill was done on November 10th, and reviled a number of small methane sources near landfill M. Southeast of the landfill was two piles of manure giving slightly elevated methane concentrations on the road close to the piles. Just next to the landfill was a chicken farm also showing slightly elevated methane concentrations as well as a cow farm approximately 1500 m east of the landfill. However, none of the three sources close by the landfill showed concentration levels that could significantly influence the quantification of methane from landfill M. The location of landfill M and the three local methane sources and the methane concentrations measured close to the individual sources are illustrated in Figure 2.

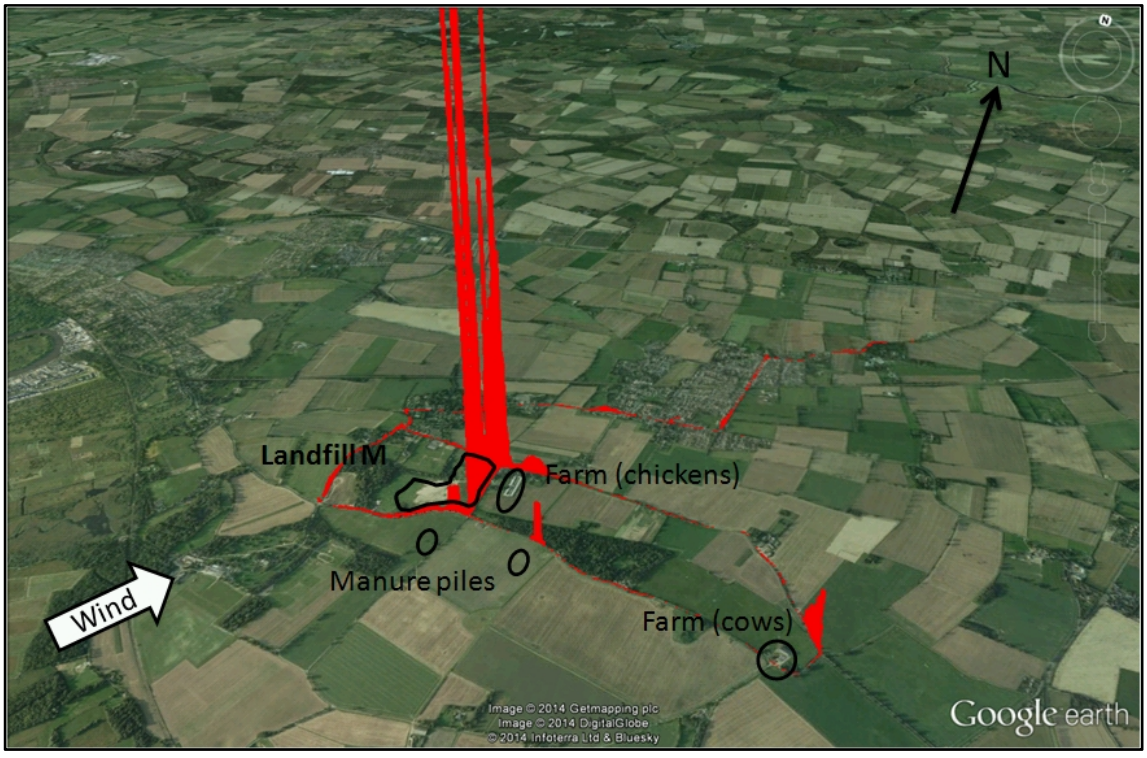

Figure 2. Atmospheric methane (red) concentrations measured in the area around landfill M. The background methane concentration is subtracted. Maximum concentrations above background were about 22 ppb at the manure piles, 27 ppb at the cow farm and 14 ppb at the chicken farm.

The methane screenings were done with wind from south and west-southwest. However, during emission quantification on November 11th, the wind came from southeast and a broad methane plume was measured downwind from the landfill. This plume was much broader than the contribution from the landfill, which indicated an additional source far from the landfill emitting a significant amount of methane. After the quantification on November 11th, an additional screening was performed far upwind from the landfill and elevated methane concentrations were found when getting closer to a sugar factory approximately 5 km southeast of landfill M. The additional screening is illustrated in Figure 3.

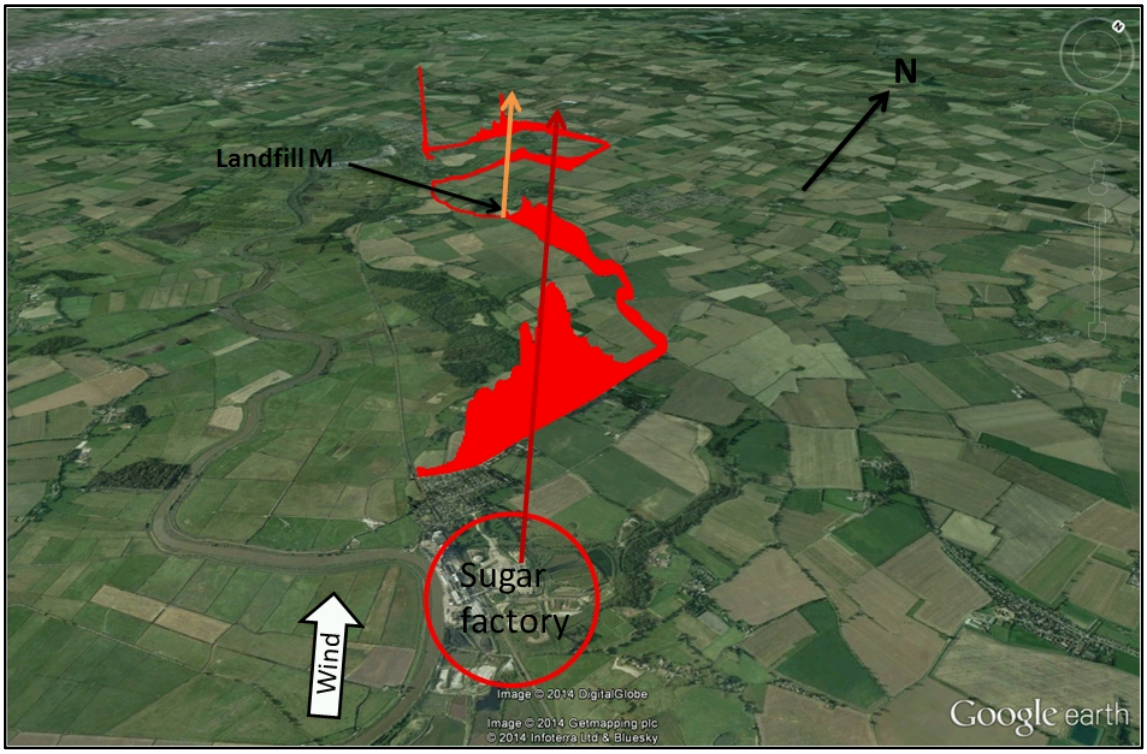

Figure 3. Atmospheric methane concentrations downwind from a sugar factory about 5 km from landfill M in the evening on November 11th. Arrows indicates the approximately plume direction of the methane from the sugar factory (red arrow) and landfill M (yellow arrow). The methane background concentration is subtracted. Maximum atmospheric concentrations above background were about 800 ppb in the plume downwind the sugar factory and about 150 ppb downwind the landfill.

6.1.2 Initial on-site or near-by methane screening of landfill M

The landfill was not accessible for onsite measurements. However, measurements close by, with a stable wind direction, indicated that the majority of the emitted methane came from the eastern landfill area, with some contribution from the middle area and little contribution from the western area. Three tracer gas bottles were used and were placed at location 1, 2 and 3 or 1, 3 and 4 in order to best simulate the methane emitted from the landfill. Figure 4 illustrates these areas and shows the chosen tracer gas placements. The initial screening of the area with a more western wind also indicated some hotspots with a larger methane emission. These hotspots were located a little north of the southeast corner, in the northeast corner and a smaller source in between those two (see Figure 5).

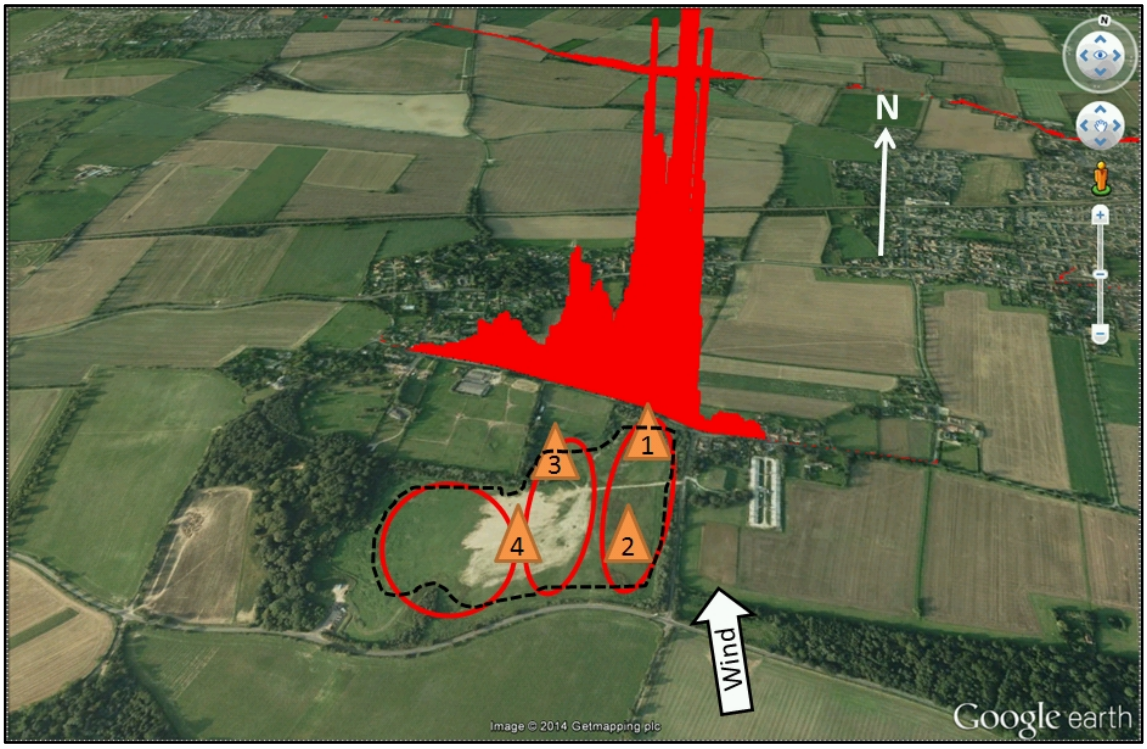

Figure 4. Atmospheric methane concentrations measured close to landfill M showing the approximate emission areas and the tracer gas placements (triangles). The dotted line marks the approximate landfill area and the red circles shows the emission areas (east, middle and west) resulting in three downwind plumes. The largest emission is seen from the eastern part of the landfill. Maximum atmospheric methane concentrations above background were about 800 ppb.

Figure 5. Screening of atmospheric methane concentrations around landfill M with wind coming from west. Maximum atmospheric methane concentrations above background were about 7500 ppb.

6.1.3 Whole landfill site methane emission from landfill M

Methane emission quantifications were done on 11th, 12th and 13th of November. The wind came from southwest on Tuesday November 11th and as explained in the screening section (6.1.1) there was some interference with a methane plume from a sugar factory approximately 5 km upwind the landfill. However, many traverses were done while driving in a square route northwest of the landfill and in several of them, it was possible to distinguish between the methane from the landfill and the methane from the sugar factory. An example of a plume measured downwind the landfill is shown in Figure 6, where it can be seen that there is a general elevated methane concentration on the roads northwest of the landfill. This elevated concentration is caused by methane emission from the sugar factory (not present on the figure, as it is 5 km upwind from the landfill location). The methane emission from the landfill can be seen as an additional increase at the same location where the tracer gas plume was measured.

Table 2 lists the methane emission based on the plume traverses conducted on November 11th. The average methane emission was 26.7±3.8 kg h^{-1}, which was the methane from the landfill alone, not including the sugar factory. The emission from the sugar factory was excluded/avoided by subtracting the background measured on each side of the plume at each transect. This is possible as the contribution from the sugar factory gives a broad and flat plume at the measurement roads due to the great distance to the factory. However, it is a plume and not a constant background concentration, which can results in a larger uncertainty in the emission quantification and open up for the possibility that a small part of the measured methane in the plume can origin from the sugar factory. Concentrations plots of two downwind plumes are shown in Figure 7. Figure 8 illustrates the background subtraction and possible contribution to the landfill plume from the sugar factory.

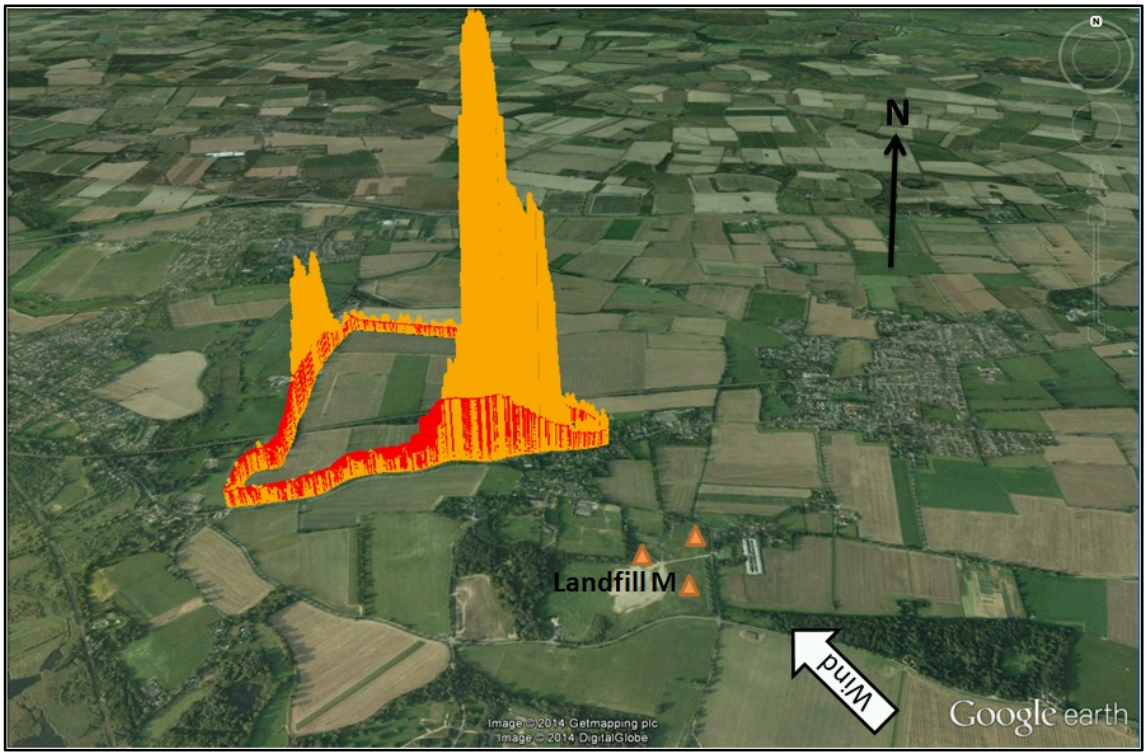

Figure 6. Methane (red) and tracer gas (yellow) concentrations at two distances (approximately 800 and 1600 m) downwind from landfill M on November 10th. Triangles illustrate the placement of the tracer gas bottles. Maximum atmospheric concentrations above background were about 260 ppb methane and 8 ppb acetylene.

Table 2. The calculated methane emission rates from landfill M calculated from each plume transect performed on November 11th 2014.

Time **(UK winter time)**	**Measured methane emission rate** **(kg h^{-1})**
18.01	30.7
18.04	22.3
18.10	23.5
18.13	29.5
18.19	23.9
18.22	23.1
18.43	21.0
19.09	35.4
19.12	30.8
Average	**26.7**
Std deviation	**5.0**
Std error of mean	**1.7**
Degree of freedom	**8**
95% confidence interval (t-distribution)	**±3.8**

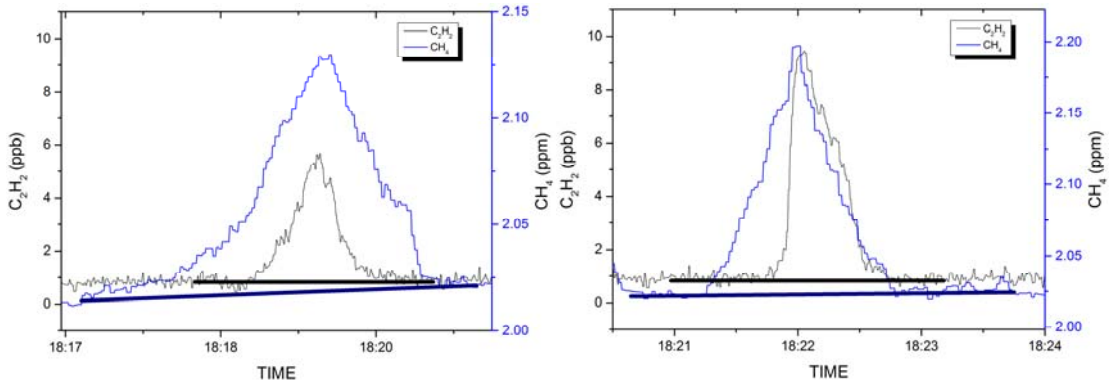

Figure 7. Methane and tracer gas plume approximately 1600 m (left) and 800 m (right) downwind from landfill M on November 11th. The thick lines show the background used for subtraction before plume integration.

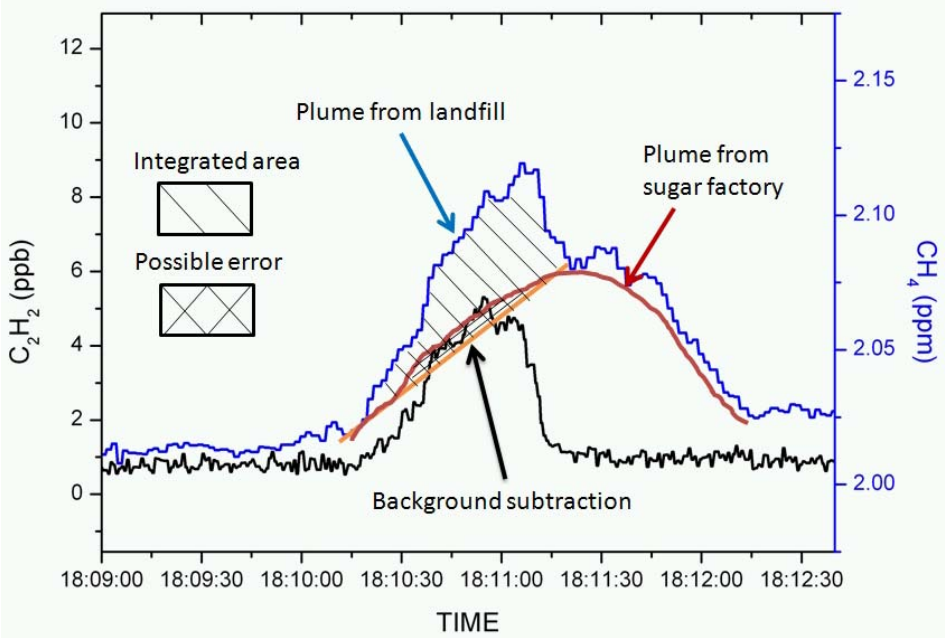

Figure 8. Methane and tracer gas plume approximately 1600 m downwind from landfill M on November 11th. Drawing shows the background subtraction and the possible error (area between orange and red curve) due to plume from the sugar factory.

On Wednesday November 12th, the wind came more from the south, which excluded any interfering methane from the sugar factory. Useful plume concentration of both methane and tracer gas was found on a road going east-west, 1500 to 2000 meters north from the landfill. Figure 9 shows the tracer gas placement on the landfill and a typical plume transect downwind on the measurement road.

Figure 9. Methane (red) and tracer gas (yellow) concentrations downwind from landfill M on November 12th. Triangles illustrate the placement of the tracer gas bottles. Maximum atmospheric concentrations above background were about 15 ppb methane and 4 ppb acetylene.

In total, 29 useful traverses were performed during the tracer gas release and the individual emission rates for each transect are shown in Table 3. The average methane emission measured was 6.6±0.5 kg h^{-1}. Figure 10 illustrates two concentrations plots of downwind plumes.

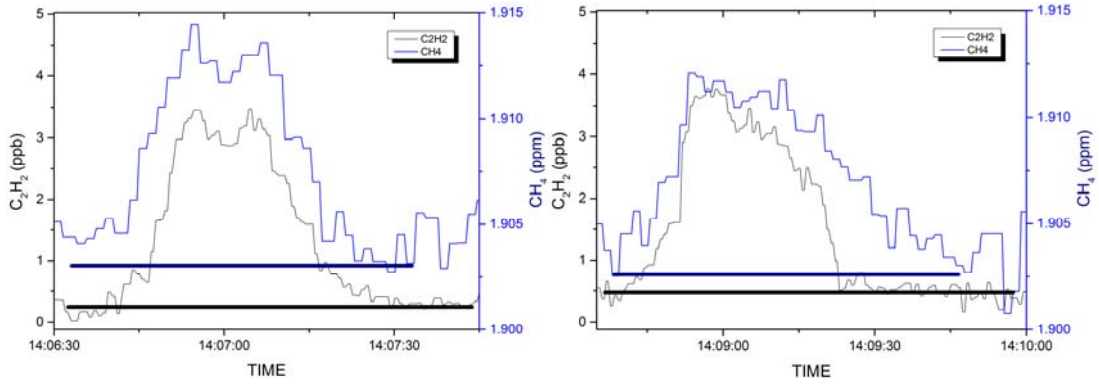

Figure 10. Two examples of methane and tracer gas plumes 1500-2000 m downwind from landfill M on November 12th. The thick lines show the background used for subtraction before plume integration.

Tabel 3. The calculated methane emission rates from each plume transect performed on November 12th 2014.

Time **(UK winter time)**	**Measured methane emission rate** **(kg h^{-1})**
12.50	7.6
13.02	7.3
13.10	8.8
13.14	7.7
13.17	8.2
13.20	4.2
13.24	5.5
13.27	5.6
13.30	6.7
13.33	6.9
13.36	6.4
13.39	5.9
13.41	4.9
13.44	6.9
13.47	6.5
13.49	8.0
13.51	6.9
13.55	6.5
13.58	6.0
14.00	6.5
14.02	7.9
14.04	8.0
14.06	6.5
14.08	5.5
14.11	9.7
14.13	6.5
14.16	5.6
14.18	4.9
14.21	4.4
Average	**6.6**
Std deviation	**1.3**
Std error of mean	**0.2**
Degree of freedom	**28**
95% confidence interval (t-distribution)	**±0.5**

On Thursday November 13th, one tracer gas bottle was moved so that the tracer gas configuration was number 1, 3 and 4 in Figure 4. Measurements were done in the morning (10 successful traverses from approx. 11.00 to 11.30 AM) and in the afternoon/evening (10 successful traverses from approx. 17.00 to 18.15). Two plume examples are illustrated in Figure 11. The two measurement periods were original done to capture the effect of using the flare at the landfill. However, the flare was taken off in the morning at 10.00 to be maintained and was not turned on later.

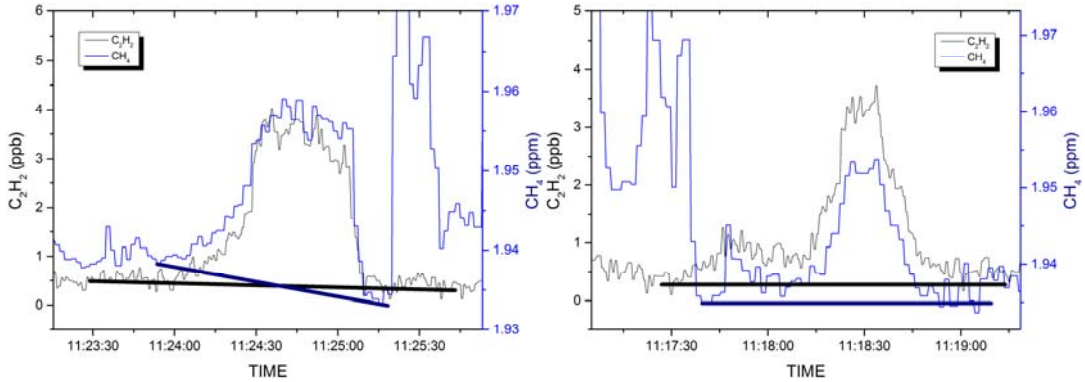

Figure 11. Examples of methane and tracer gas plumes 1500-2000 m downwind from landfill M on November 13th. The thick lines show the background used for subtraction before plume integration. Additional methane source (methane peak without associated acetylene peak) was observed close to measurement road, but did not influence the emission quantification.

Table 4 shows the measured emissions for each plume transect and the calculated average, standard deviation, standard error of mean and 95% confidence interval using the 2 sided t-distribution and the degree of freedom.

Tabel 4. The measured methane emission rates from each plume transect performed on Thursday November 13th, 2014.

Time (UK winter time)	**Measured methane emission rate ($kg\ h^{-1}$)**	**Time (UK winter time)**	**Measured methane emission rate ($kg\ h^{-1}$)**
11.07	11.2	16.57	5.9
11.11	13.6	17.02	6.9
11.14	17.1	17.28	6.0
11.16	11.7	17.34	7.2
11.18	15.6	17.39	6.7
11.20	14.8	17.44	8.2
11.22	14.6	17.53	8.0
11.25	12.4	18.01	6.8
11.27	12.7	18.07	7.8
11.29	10.8	18.14	8.6
Average	**13.5**	**Average**	**7.2**
Std deviation	**2.1**	**Std deviation**	**0.9**
Std error of mean	**0.7**	**Std error of mean**	**0.3**
Degree of freedom	**9**	**Degree of freedom**	**9**
95% confidence interval	**±1.5**	**95% confidence interval**	**±0.7**

The morning measurements gave an emission rate of 13.5±1.5 kg h^{-1} while the afternoon/evening measurements gave an emission rate of 7.2±0.7 kg h^{-1}. There were no weather changes between the two periods (rain and wind) and the atmospheric pressure was stable throughout the day.

Table 5 provides an overview over the methane emissions measured at landfill M from November 11th to 13th. The measured emission rates showed a large variation between the three measurement days. The measurements were done during a relatively large change in the atmospheric pressure, which is well-known to have an effect on the methane emission from landfills. Figure 12 shows the change in atmospheric pressure during the week of the measurements and the time of the four emission quantification measurements. During the first quantification on Tuesday (Nov. 11th) evening, the pressure was decreasing (from 1000 hPa at

12:00 to 996 hPa at 24:00), while the pressure was rapidly increasing (from 996 hPa at 8:00 to 1004 hPa at 19:00) during the measurement on Wednesday (Nov. 12th). The high decrease in atmospheric pressure during November 10th and 11th could have resulted in the high emission (26.7±3.8 kg h^{-1}) measured Tuesday evening (Nov. 11th). Around Tuesday midnight, the pressure started increasing, which could have led to a decrease in the landfill emission explaining the lower emission measured Wednesday (Nov. 12th). Furthermore, as discussed during the presentation of the results from Tuesday 11th, the higher emission rate on this day might be slightly overestimated caused by influence from the sugar factory. The pressure became stable on Thursday (Nov. 13th) and the emission was then measured to 13.5±1.5 in the morning and 7.2±0.7 in the afternoon/evening.

Another factor that could contribute to the emission variation is that the flare was burning from 8:00 to 16:00 on all days except on Thursday13th, were it was only on from 8:00 to 10:10. This means that the high emissions measured on Tuesday 11th was measured while the flare was off (measurements were done after 17:00), whereas the low emission on Wednesday 12th was measured while the flare was burning (measurements were done between 12:30 and 14:30). Information from the landfill gas manager, says that on Tuesday 11th, approximately 23 kg methane was flared per hour. If some of this methane was emitted to the atmosphere when the flare was turned off, then this could contribute to the higher emission measured in the evening on Tuesday 11th. However, the lower emissions on the 13th when the flare was off since 10:10 indicate that the atmospheric pressure might play an even bigger role on the variation. Table 5 shows an overview of the quantification measurements and the conditions.

The relatively large measurement uncertainty (14%) on Nov. 11th compared to the uncertainty during the other measurement periods (8, 8 and 10%) was due to the interference from the sugar factory, which made it more difficult to establish a background concentration to subtract from the measured downwind plumes.

Table 5. Overview over methane emissions measured at landfill M from November 11th to 13th. The table shows the change in atmospheric pressure measured over time periods of 3, 6 and 12 hours, respectively. Pressure data is from a nearby weather station. The time period is centred around the measuring time. The table also show if the flare was on or off during the measurement period. The landfill flare was burning from 8:00 to 16:00 on all days except on Thursday13th, were it was only on from 8:00 to 10:10.

Date	Time interval	Measured methane emission (kg h^{-1})	Atmospheric pressure (hPa)	Atmospheric pressure gradient 3/6/12 hours before (hPa)	Flare on or off (on or off)	Amount of methane flared (kg h^{-1})
Nov 11th	18:00 – 19:15	26.7±3.3	997.9	-0.6/-2.3/-4.1	Off (on 8:00 to 16:00)	0 (23)
Nov 12th	12:30 – 14.30	6.6±0.5	998.2	1.4/2.3/2.0	On (on 8:00 to 16:00)	23*
Nov 13th	11:00 – 11:30	13.5±1.3	1006.7	0.4/0.7/1.4	Off (on 8:00 to 10:10)	0 (23*)
Nov 13th	16:45 – 18:15	7.2±0.6	1006.0	0.0/-0.7/0.0	Off	0

*) No gas flow and concentration reported. November 10th and 11th had comparable amount of methane being flared and the same amount of methane extracted (around 23 kg h^{-1}) can thus be expected for 12th and 13th.

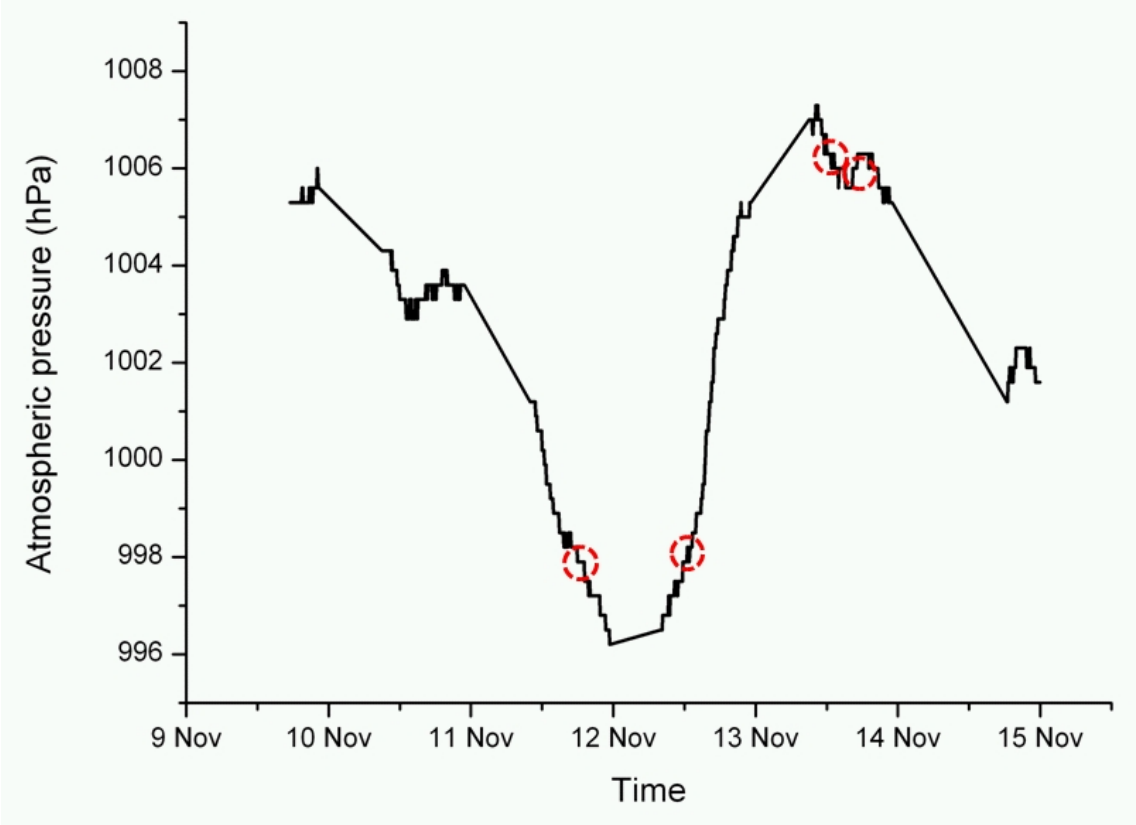

Figure 12. Atmospheric pressure during the measurement days. Red, dotted circles marks the measurement times. The data was recorded at a weather station about 10 km south of landfill M. Note that no data were available during night times, thus the partly straight line.

6.2 Landfill J

6.2.1 Methane screening of the area around landfill J

Initial screening of the area around landfill J was done on November 17th. The screening revealed a number of methane sources near the landfill. All of the sources were located at farms and are marked in Figure 13. No significant methane sources were found upwind from the landfill and the farms south and southwest of the landfill did not contribute to the downwind methane plume from the landfill. There was a small emission from a farm near the road where the landfill methane plume was located, but this small sharp peak of methane was easily recognizable and could be subtracted in the emission calculations.

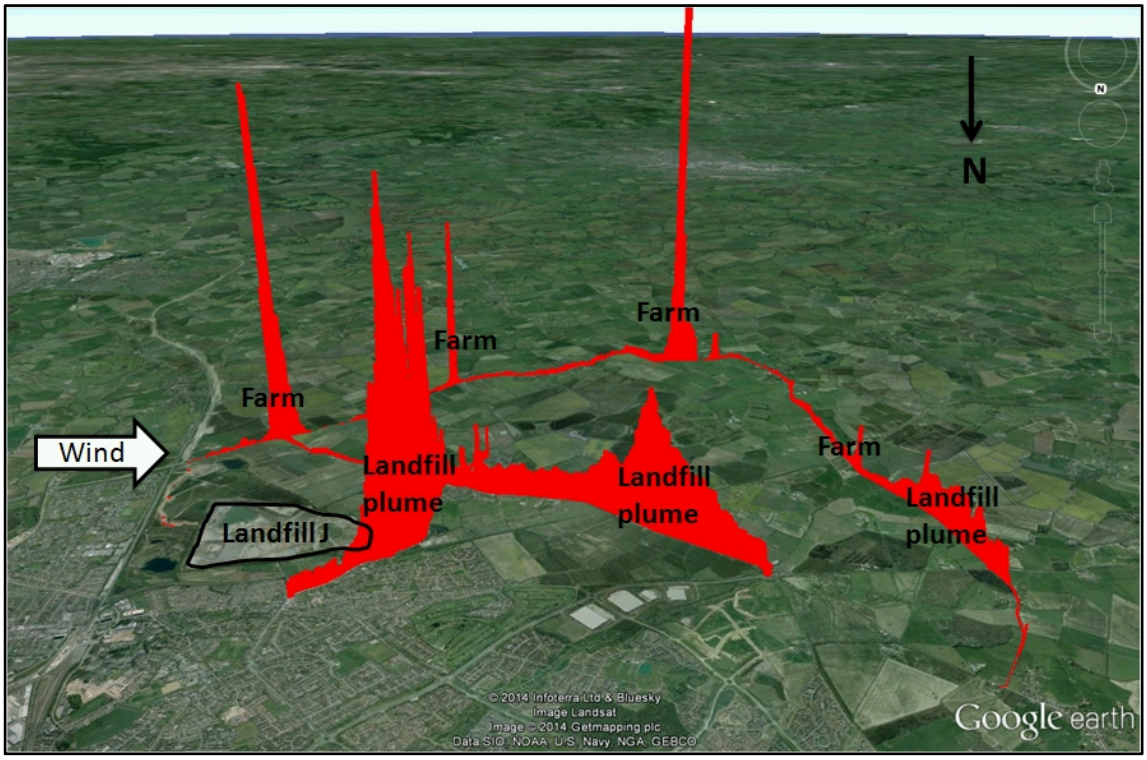

Figure 13. Relative atmospheric methane concentration around and in three distances downwind from landfill J. Background concentration is subtracted.

6.2.2 Initial on-site methane screening of landfill J

Only the main road through the landfill was accessible with the measurement vehicle. Figure 14 shows the atmospheric concentration of methane (above background concentration) on the road through the landfill. The measured concentrations indicated that methane was emitted from both the northern and southern part of the landfill. The emission from the southern part is seen where tracer gas bottle number 2 was placed and the emission from the north part is seen on the road, downwind from the landfill. It was not possible to narrow in the main emitting areas at the landfill and tracer gas bottles were placed in position 1 or 4 to insure to cover the whole landfill area. Smaller emission hotspots were also seen near the centre of the landfill and a third tracer gas bottle was placed here (location 3 on Figure 14). The valve of two of the 12 ordered tracer gas bottles were broken and the bottle in location 3 was therefore omitted on two out of the four measurement days.

Figure 14. The relative atmospheric methane concentration following the road through landfill J and on the road just downwind from the landfill. The background methane concentration is subtracted. Triangles mark the tracer gas placement used for whole site emission quantification.

6.2.3 Whole landfill site methane emission from landfill J

The previously presented Table 1 provides an overview of the tracer release and the performed measurements. Quantification of the whole landfill site emission was done on all four measurement days. Since two tracer gas bottles were broken, three bottles were used on Monday 17th and on Thursday 20th, while only two bottles were used on Tuesday 18th and on Wednesday 19th.

On the 17th, tracer gas location 1, 2 and 3 were used. The wind came from the east and the downwind plumes of methane and tracer gas were measured on a road approximately 5600 meters from the centre of the landfill. Five useful traverses were made and the resulting emission rates are shown in Table 6 and two plume examples are illustrated in Figure 15. The average methane emission was 200±48 kg h^{-1}.

Tabel 6. The calculated methane emission rates from each plume transect performed on November 17th 2014.

Time (UK winter time)	Measured methane emission rate ($kg\ h^{-1}$)
16.26	205
16.32	267
17.21	162
17.26	160
17.32	203
Average	**200**
Std deviation	**42**
Std error of mean	**19**
Degree of freedom	**4**
95% confidence interval (two side t-distribution)	**±48**

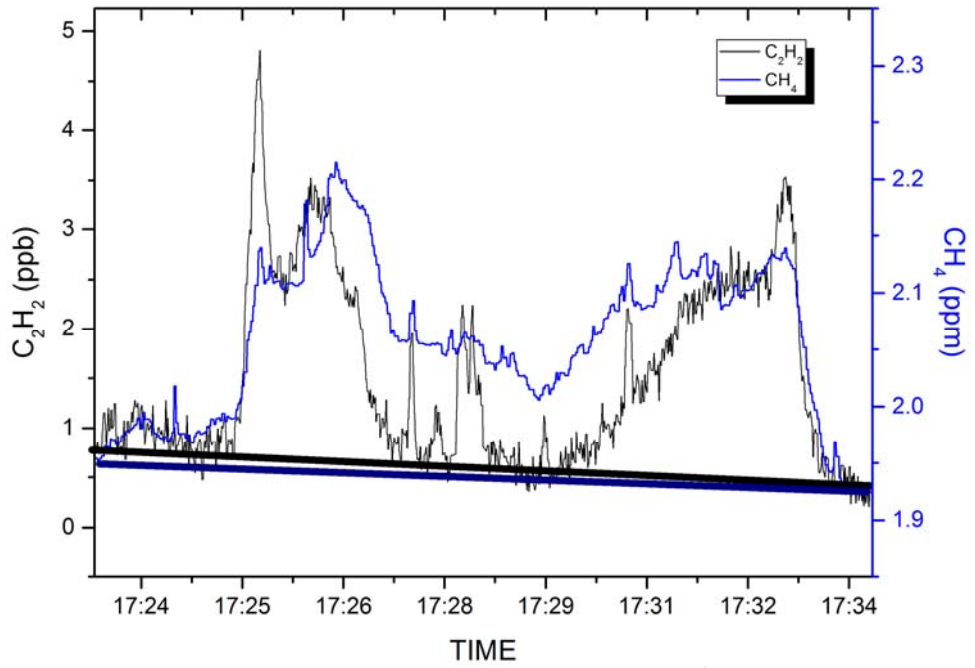

Figure 15. Examples of two plume concentration measurements on November 17th 5600 m downwind from landfill J. The thick lines illustrate the background used for subtraction before plume integration. Contributions from interfering sources were removed before integration.

On the 18th, tracer gas location 1 and 2 were used. The wind was mainly from east-northeast and the downwind plumes were more challenging to measure than the day before due to greater dispersion and lower tracer gas release rate. A total of seven useful plume traverses were performed on a road approximately 5800 meters west-southwest of the landfill, giving an average methane emission rate of 182±23 kg h^{-1}. Table 7 lists the calculated emission rates from the individual transects.

Table 7. The calculated methane emission rates from each plume transect performed on November 18th 2014.

Time (UK winter time)	Measured methane emission rate ($kg\ h^{-1}$)
16.31	181
16.40	153
16.55	188
17.01	153
17.08	198
17.13	223
17.23	175
Average	**182**
Std deviation	**25**
Std error of mean	**10**
Degree of freedom	**6**
95% confidence interval (two side t-distribution)	**±23**

The slightly different wind direction could be used to further explore the different emission areas on the landfill. Figure 16 shows the downwind plumes of methane and tracer gas including the location of the tracer gas releases. A methane plume was measured just northwest of the tracer gas plume, but not associated with the tracer gas, which indicated methane emission from the north part of the landfill. Figure 17 illustrates typical plume concentration measurements at the measurement road 5800 m downwind from the landfill. The main part of the methane plume is observed north of the tracer gas plume (the two examples in Figure 17 are when driving south to north (left) and north to south (right)), again indicating that the main emission was further north at the landfill than the tracer gas bottles.

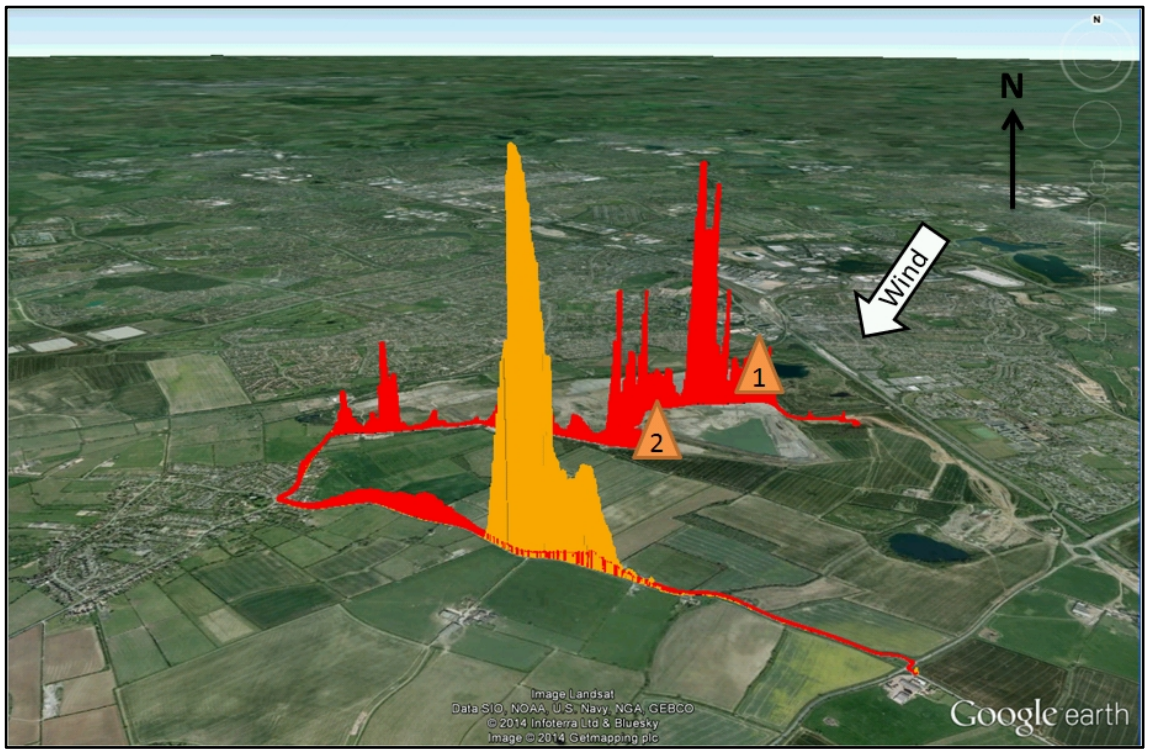

Figure 16. Downwind plumes of methane and tracer gas from landfill J on November 18th. Background concentrations are subtracted. Maximum atmospheric concentrations above background were about 9000 ppb methane at the landfill and 12 ppb acetylene.

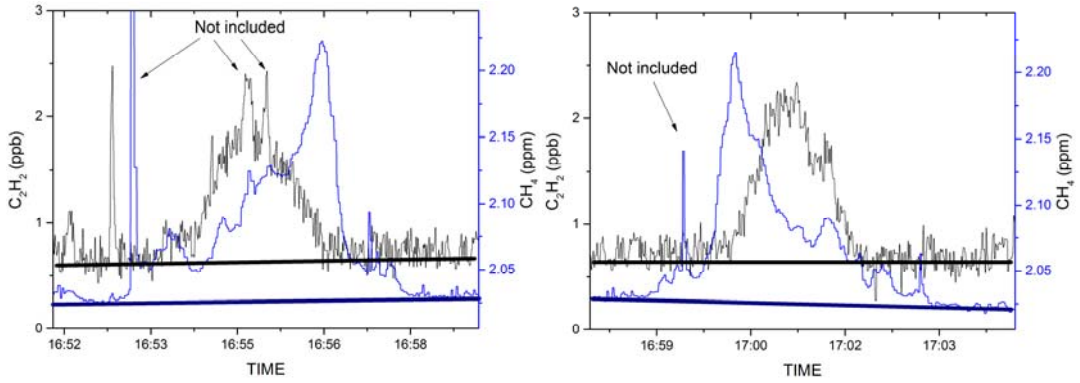

Figure 17. Examples of two plume concentration measurements on November 18th 5600 m downwind from landfill J. The thick lines illustrate the background used for subtraction before plume integration. Contributions from interfering sources were removed before integration.

On November 19th, the wind changed to come from the south-southeast and the methane plumes were measured on two roads north-northwest of the landfill. Two tracer gas bottles were placed at position 2 and 4. The wind was weak and the atmospheric dilution was large, which made the tracer gas concentrations on the measurement roads close to background concentrations. Therefore, relatively few (five) traverses were useful for quantifying the methane emission rate, which was found to be 264±61 kg h^{-1}. The individual calculated emission rates are shown in Table 8 and two plume examples are shown in Figure 18.

Tabel 8. The calculated methane emission rates from each plume transect performed on November 19th 2014.

Time **(UK winter time)**	**Measured methane emission rate** **(kg h^{-1})**
13.52	353
13.56	249
13.58	279
14.49	208
14.54	234
Average	**264**
Std deviation	**56**
Std error of mean	**25**
Degree of freedom	**4**
95% confidence interval (two side t-distribution)	**±61**

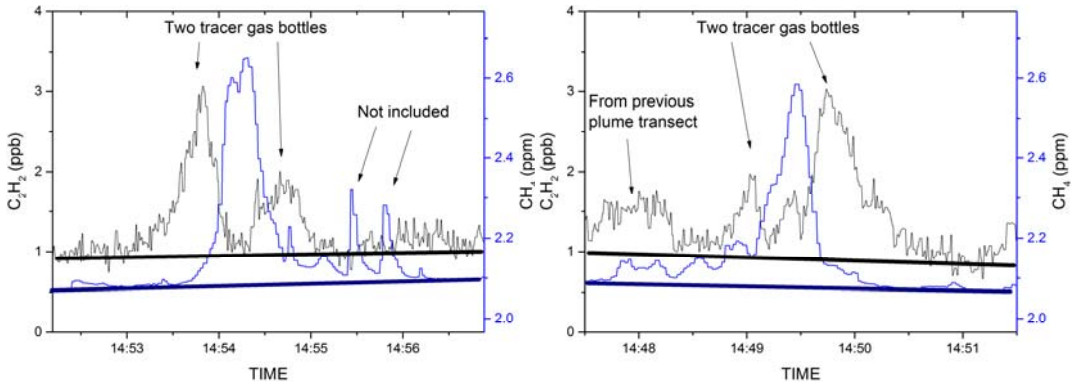

Figure 18. Examples of two plume concentration measurements on November 19th 5800 m downwind from landfill J. The thick lines illustrate the background used for subtraction before plume integration. Contributions from interfering sources were removed before integration.

On November 20th, the wind was very weak and unstable. The main wind direction in the afternoon was from the east and the downwind plumes were measured at the same road as on the first measurement day. Tracer gas position 2, 3 and 4 were used with a total tracer gas release of 2.8 kg h^{-1}. The emission from a farm nearby was easily separated (see Figure 19) from the landfill methane plume. An average methane emission of 274±33 kg h^{-1} was measured using eight successful traverses. The emission rates from the individual traverses are listed in Table 9 and examples of downwind plume concentrations are illustrated in Figure 20.

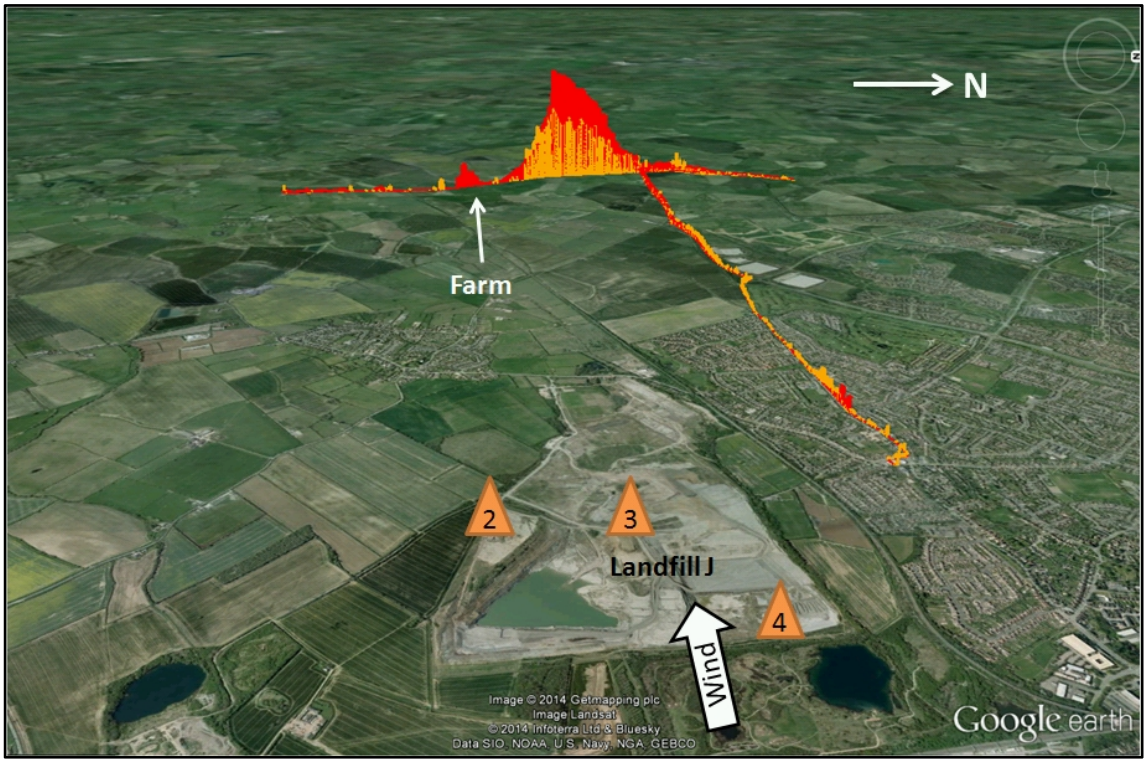

Figure 19. Tracer gas position at landfill J on November 20th and the downwind plume of methane and tracer gas, approximately 5 km away. Background concentrations are subtracted. Maximum atmospheric concentrations above background were about 300 ppb methane and 2 ppb acetylene.

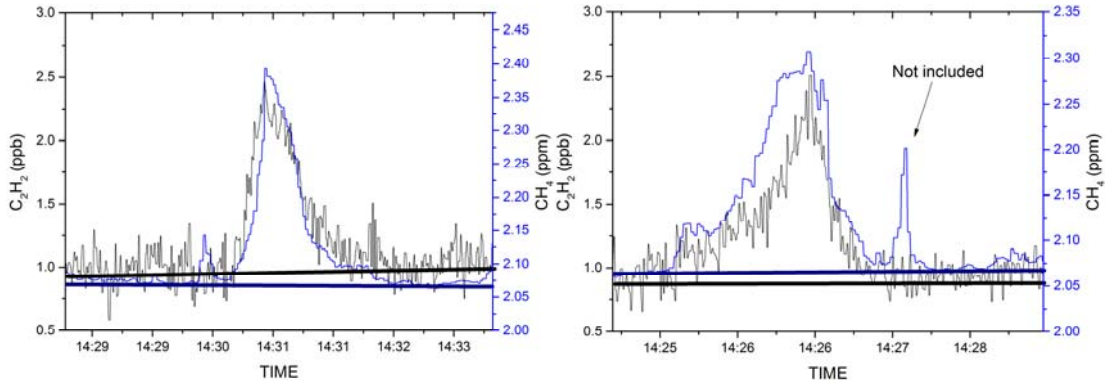

Figure 20. Examples of two plume concentration measurements on November 20th 5600 m downwind from landfill J. Thick lines illustrates the background used for subtraction before plume integration. Contribution from Interfering sources were removed before integration.

Tabel 9. The calculated methane emission rates from each plume transect performed on November 20th 2014.

Time (UK winter time)	**Measured methane emission rate ($kg\ h^{-1}$)**
14.26	307
14.31	250
14.35	320
14.38	305
14.42	306
14.45	232
14.50	208
14.53	263
Average	**274**
Std deviation	**41**
Std error of mean	**15**
Degree of freedom	**7**
95% confidence interval (two side t-distribution)	**33**

Table 10 gives an overview of the methane emission rates measured on the four different days. Also listed in Table 10 is the atmospheric pressure during the measurements and the change in the pressure 3, 6 and 12 hours prior to the measurement. For comparison, also the amount of recovered methane is listed.

Table 10. Overview over methane emissions measured at landfill J from November 17th to 20th. The table shows the change in atmospheric pressure measured over periods of 3, 6 and 12 hours before the measurements. The table also show gas collection.

Date	Time interval	Measured methane emission ($kg\ h^{-1}$)	Atmospheric pressure (hPa)	Atmospheric pressure gradient 3/6/12 hours before (hPa)	Amount of methane collected ($kg\ h^{-1}$)
Nov 17th	16:30 – 17:30	200±48	991.0	NA/NA/NA*	2064
Nov 18th	16:30 – 17.30	182±23	1000.1	+0.8/+0.9/+3.5	2075
Nov 19th	13:30 – 15:00	264±61	1007.6	0.0/+2.3/+4.3	2031
Nov 20th	14:15 – 15:00	274±33	1013.0	-1.1/-0.1/+0.6	2106

*) The atmospheric pressure was measured by the NPL from the afternoon on Nov. 17th, and thus there are no data from the hours before. Weather data was obtained from a weather station nearby shows same pressure change as the NPL, although the absolute pressure is different. The data from the external weather station showed an increase of 2.0/4.4/7.1 from 3/6/12 hours before.

In general, during the measurements at landfill J, the methane emission rate changed from day to day, but overlapping within the 95% confidence interval. Landfill emissions are influenced by

changes in the atmospheric conditions such as atmospheric pressure changes and changes in wind speeds. This is well documented in the literature and depending on the design and operation of the landfill, even small changes in the atmospheric pressure can result in relative large changes in landfill methane fluxes. Also fluctuations in the gas recovery can affect the methane emission rate. Figure 20 shows the measured methane emission rates in comparison with the atmospheric pressure. During the first three measurement days (November 17th to 19th), the atmospheric pressure was increasing towards a maximum in the pressure just before midday on the last measurement day, November 20th. Hereafter, the pressure dropped approximately 1.5 hPa between 1014 and 1012.5 hPa and became stable the rest of the day. The first three measurements were thus taken during a pressure increase, which often will dampen the emission, whereas the highest emission was measured the last day during a period with a small, but fast pressure drop, likely to overestimate the emission rate.

In theory, the tracer dispersion measurement method is not influenced by the weather conditions as long as the tracer release simulates the methane emission from the landfill. However, during the measurement days, the wind direction and the wind speed changed. If the tracer coverage of the landfill is insufficient and the measurements are performed too close to the landfill, a change in wind direction can affect the measured emission rates as the ratio between the tracer and the methane will change. However, at Landfill J the measurements were performed very far downwind from the site (about 5600m) and the possibility of that the observed variations in the emission should be caused solely by changes in wind speed is very limited. This was assessed by comparing the measuring distances between the individual tracer release points and the plume traverses conducted during times with different wind directions. On November 20th, the wind direction changed significantly during the measurement campaign (from 14:15 to 15:30). The last transects performed during this campaign (15:00 to 15:30) were excluded as the tracer simulation of the source during this period where the wind direction had changed were less optimal and could have affected the results (see conclusion). A change in wind speed will affect the travel time of the tracer and also the mixing of the tracer and methane. More stable atmospheric conditions results in less mixing of methane and trace gas, but it also allows measurements further from the emission source without reaching the detection limit of the analytical instruments.

Landfill J had gas engines utilizing the extracted landfill gas, as well as a flare. The gas engines and the stacks were located in the western part of the landfill area. A possible emission from the stacks would be emitted at an elevated height (about 20 m) and thus have a different transportation path in the atmosphere from the landfill to the measurement road in comparison to methane emitted from the landfill surfaces. To check the possible error of this vertically elevated emission on the total emission rate, calculations were done using a Gaussian plume equation. The parameters used were wind speed (3 m s^{-1}), stability class (D), emission height (20 m) and rural area. The stability class D was chosen, as the conditions best described this class, and because the measured tracer gas dilution matched this stability class. The calculations showed that 5 km from the source, the measurements would include the emission from the stacks, but underestimate the measured emission by 2%. As the total site emission was thought to be much higher than the emission from the stacks, this potential error on the total emission due to the stack emission becomes significantly smaller than 2%.

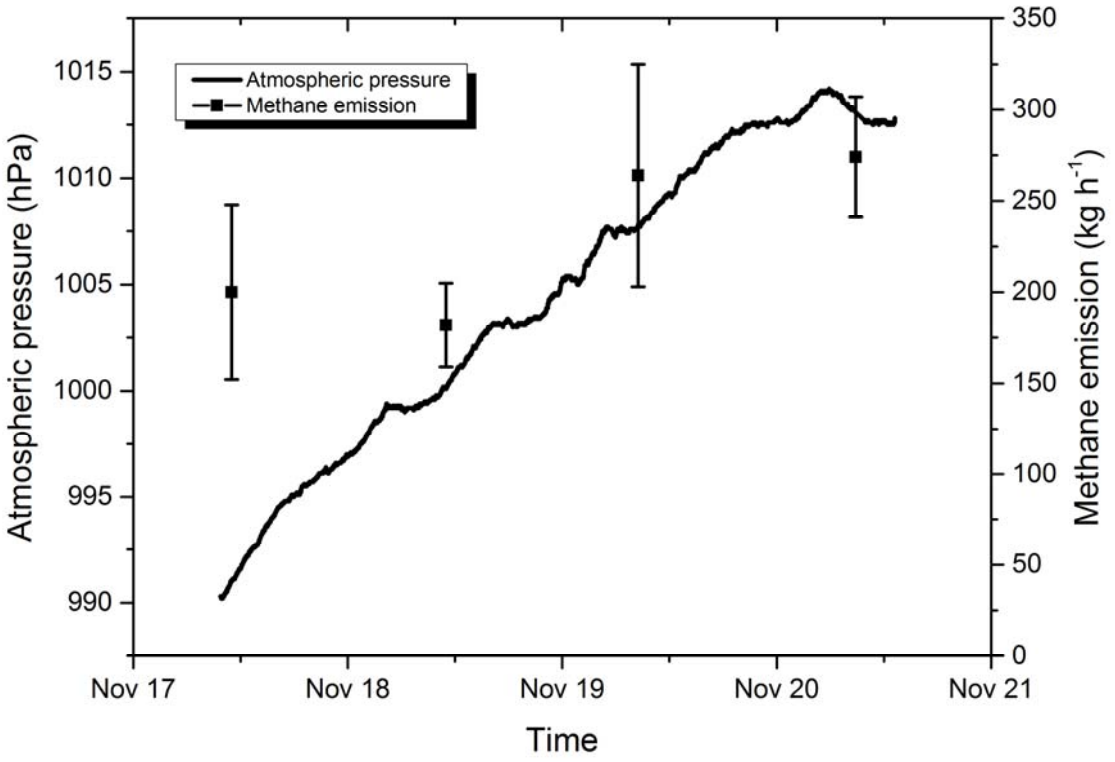

Figure 21. The atmospheric pressure during the measurement period at landfill J and the measured methane emission rates including 95% confidence interval using t-distribution. The data was recorded at a weather station about 5 km northwest of landfill J.

Each of the traverses represents the measured total site emission at the specific time of the measurement. The overall emission from landfill J during the four days can be calculated as the average of all the traverses, assuming that they each represent an equal time of emission and that these emissions were representative for the time where no measurements were performed. The overall average emission rate is then 231±23 kg h^{-1}, where the uncertainty is the 95% confidence interval of a two-sided t-distribution using the N-1 as the degree of freedom.

6.3 Landfill N

6.3.1 Methane screening of the area around landfill N

The conditions and tracer gas release can be seen in the previously presented Table 1. An initial screening of the atmospheric methane concentrations was done in the area around landfill N. Figure 22 shows the methane concentration above the background methane concentration. Besides landfill N, only the older landfill to the north had significant methane emissions. A smaller unidentified methane source was found east of the landfill, but the methane plume from this source did not overlap with the methane plume from landfill N, but rather with the methane plume from the old landfill. No elevated methane concentrations were seen at the road east of the two landfills, which indicate that the unknown source did not contribute to the plumes measured two kilometres downwind.

Figure 22. The methane concentration in the area around landfill N. Background concentration of 1998 ppb was subtracted.

6.3.2 Initial on-site methane screening of landfill N

The road conditions at the landfill did not allow the measurement vehicle to drive on site and therefore no onsite screening was performed. The placement of tracer gas bottles (see Figure 23) was chosen after conversation with the landfill operators about suspected emission areas. Two bottles were placed at the middle of the landfill, next to a large landfill area temporary covered with plastic. During the placement, landfill gas could be smelled. A third tracer gas bottle was placed in the north part of the landfill to mark the edge between the measured landfill and the older landfill to the north. A forth tracer gas bottle was placed at the south part of the landfill to mark the edge of the landfill in that direction, perpendicular to the wind direction. Each of the four bottles was set to release 928 gram per hour.

Figure 23. Placement of the tracer gas bottles (orange triangle) at landfill N on November 21th.

6.3.3 Whole landfill site methane emission from landfill N

The wind from east-southeast allowed plume measurements at a road 2000 to 2500 meters downwind from landfill N. The road ran through a town and the traffic complicated the measurements, as traffic lights and queue made it difficult to measure at a constant driving speed. The measured concentrations were therefore plotted as a function of meters traversed (instead of time, which can only be used at relatively constant driving speed) before the plumes were integrated. Figure 24 shows the methane and tracer gas plumes downwind from the landfill. A total of 12 successful plume transects were performed and gave an average emission rate of 523±60 (95% confidence interval). The calculated emission rate from the individual transects are listed in Table 11. Each emission rate was calculated by integrating the plume and finding the tracer gas/methane ratio. Two examples of such plumes are showed in Figure 25, where is can be seen that the tracer gas from the bottle placed on the north side of landfill N follows the methane very well while the tracer gas bottle marking the south edge of the landfill was placed a little too far south compared to the main emission from the site. The emission was therefore slightly more concentrated in the mid and northern parts, but nicely separated from the emission from the other landfill.

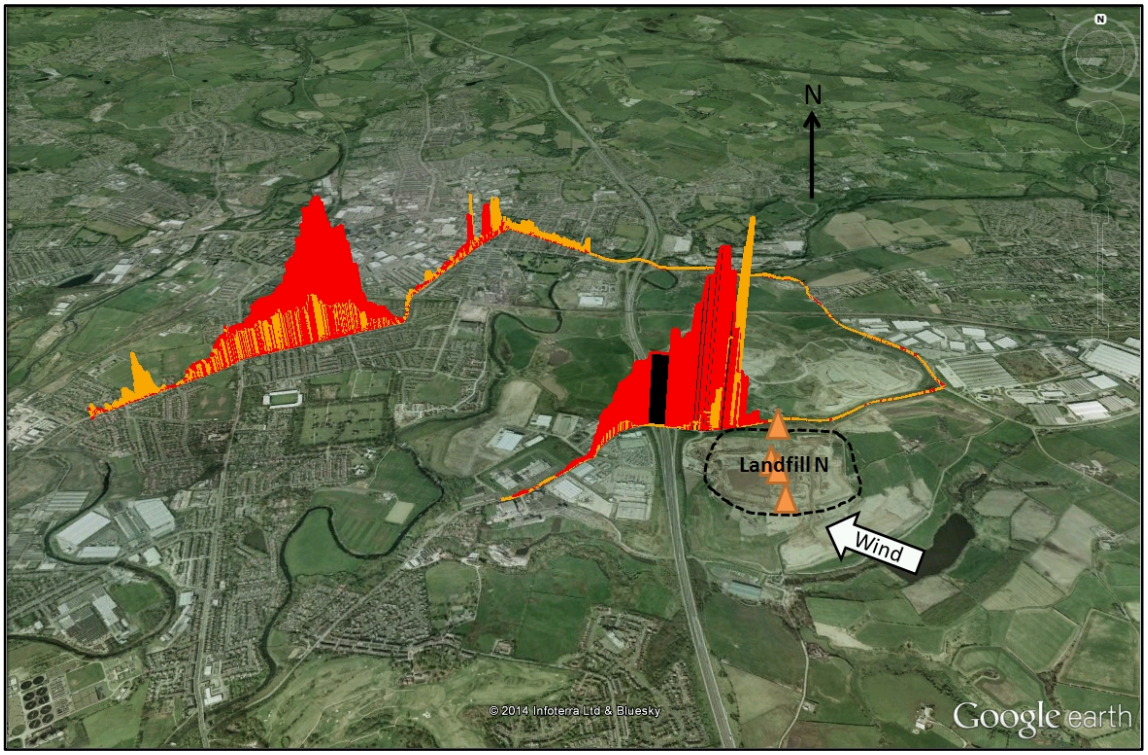

Figure 24. Atmospheric concentrations of methane (red) and tracer gas (orange) at two roads downwind from landfill N during tracer gas release. Background concentrations of 1998 ppb methane and 0.6 ppb tracer gas are subtracted. Maximum atmospheric concentrations above background were about 8000 ppb methane and 180 ppb acetylene and the road close by and about 670 ppb methane and 2.5 ppb acetylene and the road 2 km downwind.

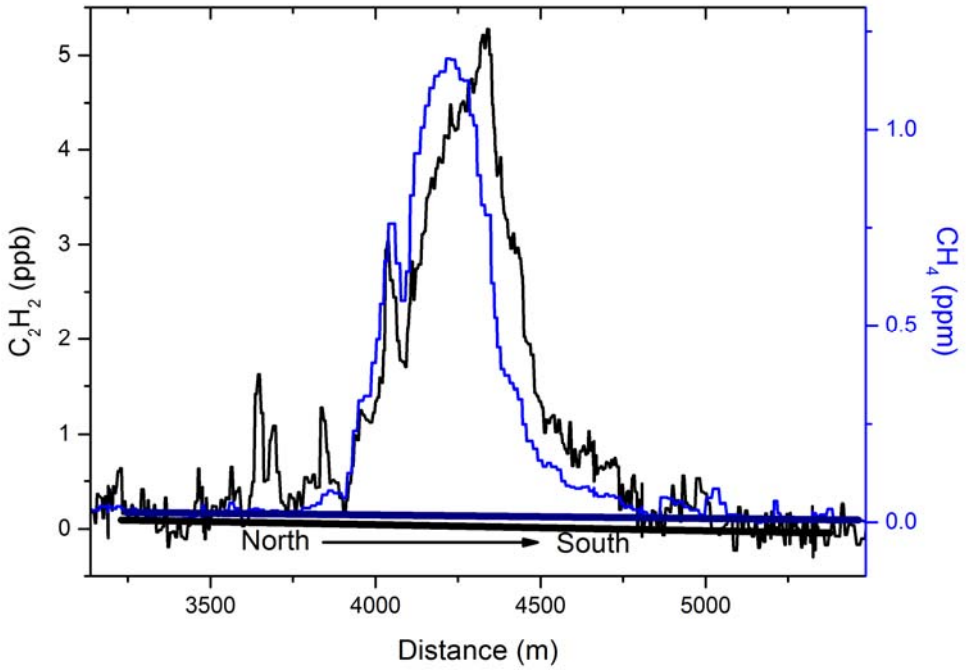

Figure 25. Example of the methane and acetylene concentrations downwind, on the road 2000-2500 m west of landfill N. The thick lines illustrate the background which was subtracted before integration of the plumes. Measurements were done in a town with many stops due to traffic and the x-axe is therefore transformed into distance instead of time.

Table 11. The calculated methane emission rates from each plume transect performed on November 21th 2014.

Time **(UK winter time)**	**Measured methane emission rate** **(kg h^{-1})**
13.50	405.6
13.56	408.0
14.08	483.1
14.16	435.5
14.20	432.7
14.25	622.7
14.35	505.9
14.40	644.4
14.45	588.0
14.51	612.2
14.57	642.3
15.03	497.9
Average	**523.2**
Std deviation	**93.6**
Std error of mean	**27.0**
Degree of freedom	**11**
95% confidence interval (2 sided t-distribution)	**±59.5**

The older landfill just north of landfill N also emitted methane. Quantification of the emission was not a part of the plan and thus no tracer gas bottles were placed there. However, the wind direction and measurement road made the distance from the two landfills to the measurement road approximately the same and a rough emission rate estimate from the old landfill can be made using the dilution factor from landfill N. Figure 26 shows the two downwind plumes from the two landfills. The transect done for estimation of the methane emission from the old landfill was done by continuing on the south-north road, rather than turning towards east like in the example in Figure 26. Two integrated plumes from the old landfill showed a methane emission rate of 25 and 34 kg h^{-1}, respectively, which can be seen as a rough estimate.

Table 12. Overview over methane emissions measured at landfill N November 21st. The table shows the change in atmospheric pressure measured over time periods of 3, 6 and 12 hours before the measurements. The table also show gas collection.

Date	Time interval	Measured methane emission (kg h^{-1})	Atmospheric pressure (hPa)	Atmospheric pressure gradient 3/6/12 hours before (hPa)	Amount of methane collected (kg h^{-1})
Nov 21st	13:50 – 15:10	523±60	1018.3	-3.3/-5.6/-8.3	956

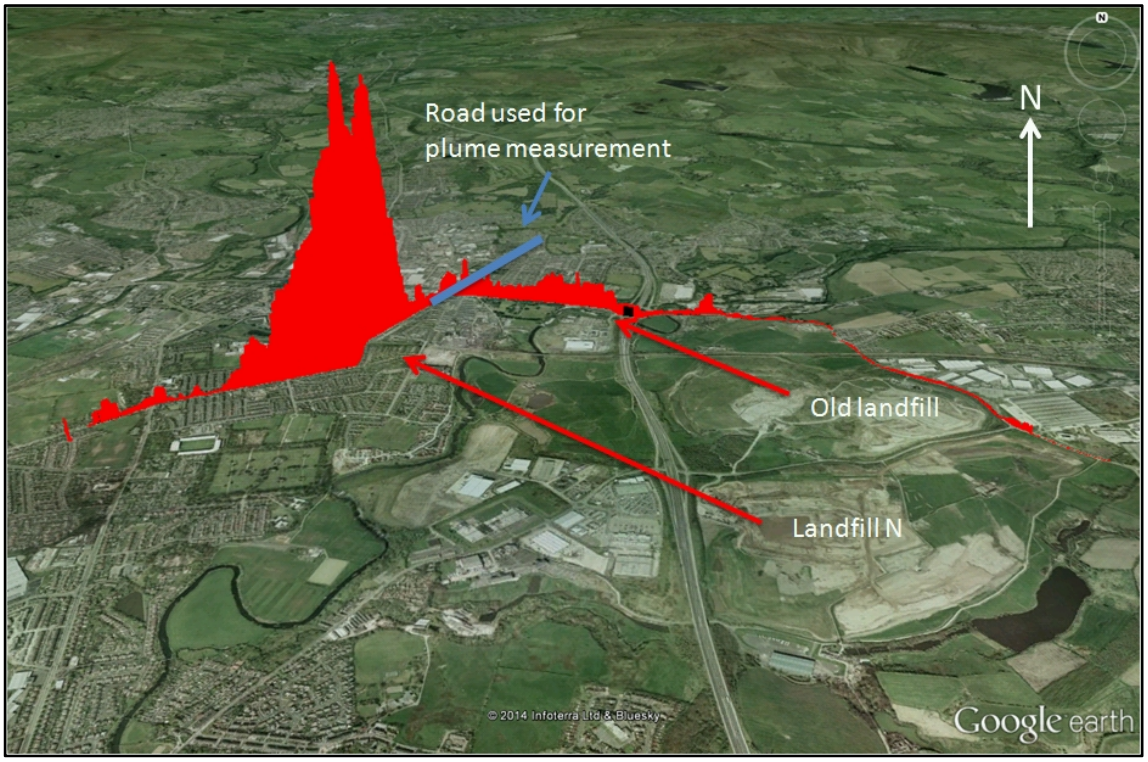

Figure 26. Methane plumes downwind from landfill N and the older landfill just north of landfill N. Background concentrations are subtracted. Maximum concentrations measured were 650 ppb methane.

6.4 Emission from all three landfills

The methane emission rates from all three UK landfills, measured in November 2014 using the tracer dispersion method are listed in Table 12.

Table 12. All measured methane emission rates at the three UK landfills measured in November 2014.

Landfill	Date	Measuring time interval	Number of traverses	Measured methane emission (kg h^{-1})	Methane recovered (kg h^{-1})	Atmospheric pressure/ gradient in 6 hours (hPa)
M	Nov 11	18:00 – 19:15	9	26.7±3.3	0	997.9/-2.3
	Nov 12	12:30 – 14.30	29	6.6±0.5	25	998.2/+2.3
	Nov 13	11:00 – 11:30 & 16:45 – 18:15	10 & 10	13.5±1.3 & 7.2±0.6	0	1006.7/+0.7 1006.0/-0.7
J	Nov 17	16:30 – 17:30	5	200±48	86.0	999.2/+4.4
	Nov 18	16:30 – 17:30	7	182±23	86.4	1009.4/+1.7
	Nov 19	13:30 – 15:00	5	264±61	84.6	1017.2/+2.1
	Nov 20	14:15 – 15:30	8	274±33	87.7	1021.9/-1.7
N	Nov 21	13:50 – 15:10	12	523±59	956	1018.3/-5.6

7. Discussion

The methane emission rates from three UK landfills were successfully quantified using the tracer dispersion method. There was a large difference in methane emissions from the three different landfills with lowest emission of 6.6±0.5 kg h^{-1} measured at landfill M and highest emission of 523±59 kg h^{-1} measured at landfill N. The difference in methane emissions between landfill sites

is a result of different waste amounts, waste compositions, waste disposal age, landfill designs (membranes, gas collection, cover, etc.). Variations in emissions between measuring campaigns performed at individual sites were also seen. Emission variations over time frames of hours or days are expected due to influence of barometric pressure, wind speed, changes in gas collection rates, etc. However, at some landfills significant variation in the methane emission was observed between individual plume transects performed 15-30 minutes after each other. These short term emission changes are more difficult to explain. In the following section different factors, which potentially could influence the true methane emission rate as well as factors that could influence the measuring method resulting in emission variation is explored.

Wind direction change and source simulation

The atmospheric pressure was unstable throughout the two measurement weeks, which makes it a challenge to estimate an average emission rate able to upscale to a yearly emission from the individual landfill. The measurements performed on November 20th at landfill J showed the importance of correct tracer gas configuration and corresponding measurements at suitable roads. On November 20th, the measured methane emission rate changed during the one hour of measurement during a period where also the wind direction and speed changed. There can be several reasons for change in methane emission during this period. One reason could be an actual change in the emission due to changes in atmospheric conditions (such as atmospheric pressure and wind speed). Such emission changes have previously been reported in the literature, and the magnitude and time response of these changes are highly site specific. Another reason could be an artefact in the measurements. Figure 27 shows the measured methane plumes approximately 5600 m west of landfill J on November 20th. The last two plume transects were measured after a change in wind direction (and wind speed) and resulted in lower measured emission rates. The lower methane emission rate could be a result of an incorrect source simulation as the tracer gas bottles were placed too far in the southwest corner of the landfill compared to the main emission areas after the wind direction had changed. In this situation, the tracer gas would then be overestimated at the measurement road and the methane would therefore be underestimated. This shows the importance of a good site screening of especially large landfills (such as landfill J) for obtaining accurate emission areas for tracer gas placement. The last two transect performed on November 20th were not included in the emission calculations as presented in the earlier sections of the report.

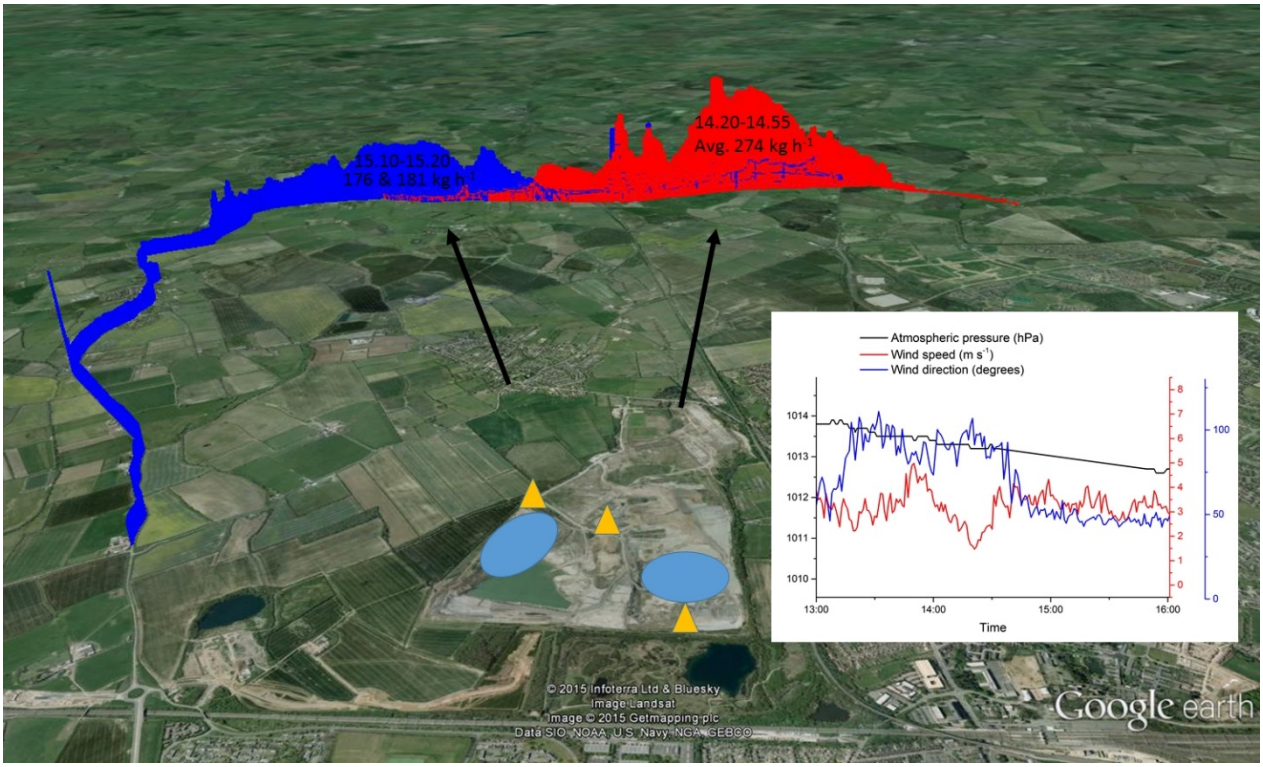

Figure 27. Methane plumes downwind from landfill J. Background concentrations are subtracted. Red shows all plumes measured before the wind direction and speed changed and the blue shows the two plumes measured after wind change. The orange triangles mark the tracer gas placement and the light blue circles marks the main emission areas found by NPL. Inserted graph shows the meteorological data measured by NPL. Note the shift in time between the recorded wind change and the time for the measured plumes 56000 m downwind from the landfill. This delay corresponds to the time for the emitted gasses to travel from the landfill to the measurement road with the current wind speed.

Data quality assessment using the tracer to methane plume concentration correlation coefficient

The methane emission is calculated based on the methane to tracer ratio, which can be obtained in different ways. Mønster et al., (2014) tested four approaches to obtain the methane to tracer ratio using the data from a large-scale controlled release test. The first approach applied a fully integration of both plumes to obtain the methane to tracer ratio, the second approach used the peak height of the plumes, the third used a scatter plot of the concentrations, and finally the last approach included a Gaussian fitting of the methane plume and subsequent inverse modelling. In an ideal situation with a point source giving perfect mixing of methane and tracer gas, undisturbed Gaussian distribution of the plume and known atmospheric stability parameters, all four approaches would yield the same emission result. The results presented in Mønster et al. (2014), showed that the plume integration is the best approach and significant over- and underestimation of the emission rate can occur using the other approaches especially when the tracer gas bottles are misplaced compared to the methane emission area. In this study, the methane to tracer ratio was obtained by full-integration of both the methane and the tracer plume.

However, one potential advantage of the using the scatter plot of methane to tracer concentrations measured in the plume is that correlation cut-off values for the inclusion of a traverse could be established and used for quality assessment. Recent literature has suggested using the methane to tracer concentration correlation coefficient (r^2) for each individual plume traverse as a selection/quality parameter. Roscioli et al. (2015) suggested only using plume traverses where the methane to tracer correlation has a $r^2 > 0.75$ whereas Foster-Witting et al. (2015) suggested using $r^2 > 0.80$ as a quality parameter, as well as visual selection and a signal-to-noise ratio > 10. The

application of cut-off values has been suggested as a way for non-specialist to perform data processing and emission calculations.

In this study, the usefulness of each plume traverse was assessed by visual inspection and selection using the knowledge about the possible methane sources in the area that could influence the methane plume from the landfill. The influence on the obtained methane emission rate of acceptance of plume traverses based on visual inspection (as done is this study) and by applying cut-off values for methane to tracer scatter plots was assessed by comparing the average emission obtained when applying these two approaches. Two data sets from landfill M were chosen for the comparison. Table 13 provides an overview of the effect on the average methane emission rate when applying r^2 cut-off values to the plume traverses measured at the landfill M. Applying cut-off values to the data, resulted in the exclusion of some of the first transects in the measurement series. The average methane emission rate for the measurement series however remained approximately the same (originally 13.5 kg h^{-1} in comparison to 13.2 kg h^{-1} and 13.4 kg h^{-1}, when applying a r^2 of 0.75 and 0.8, respectively). The confidence interval increased due to the lower number of useful traverses, causing less degrees of freedom and a larger t-value.

Table 13. Overview of the effect on the average methane emission when the applying cut-off values for the tracer/methane correlation coefficient to the plume traverses measured at the landfill M, Nov. 13th during 11:00 to 11:30.

Measurement time (centre of plume)	Measured methane emission rate (kg h^{-1})	Tracer/methane correlation coefficient (r^2)	Measured methane emissions rate included when applying the cut-off values for r^2	
	Visual selection	r^2	$r^2 > 0.75$	$r^2 > 0.80$
11.07	11.2	0.64	-	-
11.11	13.6	0.58	-	-
11.14	17.1	0.63	-	-
11.16	11.7	0.79	11.7	-
11.18	15.6	0.95	15.6	15.6
11.20	14.8	0.78	14.8	-
11.22	14.6	0.92	14.6	14.6
11.25	12.4	0.92	12.4	12.4
11.27	12.7	0.77	12.7	-
11.29	10.8	0.85	10.8	10.8
Average	**13.5**		**13.2**	**13.4**
Std deviation	**2.1**		**1.8**	**2.2**
Std error of mean	**0.7**		**0.7**	**1.1**
Degree of freedom	**9**		**6**	**3**
95% confidence interval	**±1.5**		**±1.7**	**±3.4**

Also for the other tested data set (Table 14), the three approaches give very similar results.

Table 14. Overview of the effect on the average methane emission when the applying cut-off values for the tracer/methane correlation coefficient to the plume traverses measured at landfill M, Nov. 12th during 12:30 to 14:30.

Measurement time (centre of plume)	Measured methane emission rate ($kg\ h^{-1}$)	Tracer/methane correlation coefficient (r^2)	Measured methane emissions rate included when applying the cut-off values for r^2	
	Visual selection	r^2	$r^2 > 0.75$	$r^2 > 0.80$
12.50	7.6	0.55	-	-
13.02	7.3	0.83	7.3	7.3
13.10	8.8	0.44	-	-
13.14	7.7	0.70	-	-
13.17	8.2	0.72	-	-
13.20	4.2	0.87	4.2	4.2
13.24	5.5	0.62	-	-
13.27	5.6	0.83	5.6	5.6
13.30	6.7	0.66	-	-
13.33	6.9	0.75	6.9	-
13.36	6.4	0.78	6.4	-
13.39	5.9	0.75	5.9	-
13.41	4.9	0.73	-	-
13.44	6.9	0.83	6.9	6.9
13.47	6.5	0.83	6.5	6.5
13.49	8.0	0.80	8.0	8.0
13.51	6.9	0.86	6.9	6.9
13.55	6.5	0.87	6.5	6.5
13.58	6.0	0.74	-	-
14.00	6.5	0.83	6.5	6.5
14.02	7.9	0.82	7.9	7.9
14.04	8.0	0.77	8.0	-
14.06	6.5	0.90	6.5	6.5
14.08	5.5	0.84	5.5	5.5
14.11	9.7	0.81	9.7	9.7
14.13	6.5	0.80	6.5	6.5
14.16	5.6	0.81	5.6	5.6
14.18	4.9	0.64	-	-
14.21	4.4	0.88	4.4	4.4
Average	**6.6**		**6.6**	**6.5**
Std deviation	**1.3**		**1.3**	**1.4**
Std error of mean	**0.2**		**0.3**	**0.3**
Degree of freedom	**28**		**19**	**15**
95% confidence interval (t-distribution)	**±0.5**		**±0.6**	**±0.7**

For the two larger landfills (landfill J and N), the methane to tracer correlation coefficients would be lower, and most likely also lower than 0.75 (not tested by calculations). The main reason for this was the poorer coverage of the landfill methane emission by the tracer gas bottles at these landfills. However, the methane emission can still be estimated from these landfills by integrating the area under the two plumes and taking the ratio of these plume areas.

Choice of methane background concentration

To calculate the methane emission, the plume concentrations need to be adjusted by subtraction of the background concentrations of both methane and the tracer gas (in this case acetylene). The background concentration of methane can change relatively fast if the atmospheric conditions changes. This has been seen in several studies. This change is usually caused by a change in solar influx and thus a change in the thickness of the boundary layer. When relatively strong methane sources are present in the area, a decrease of the boundary layer thickness would result in a higher background concentration and vice versa. In addition to being time dependent, a change in

background concentration can be location dependent. In such a case, methane plumes can have higher or lower background concentration on each side of the plume. This is caused by an additional methane source influencing the plume. Usually, one try to avoid such situations by choosing an appropriate wind direction and measurement road where other sources do not interfere, but unforeseen sources and/or wind direction changes can occur. An example of this was seen in landfill M where a sugar factory influenced the downwind methane concentration at a certain wind direction present on the first day of quantification. When this is observed during the measurements, then the transect was continued to ensure that the additional methane was not coming from the landfill or a source closer to the measurement road (the background concentration would then fall back to "normal" level if that was the case). In the case of the sugar factory, the higher background stayed many hundred meters after the landfill plume had ended and then slowly decreased, indicating a relatively strong methane source much further away than the landfill.

When the methane background is not influenced by changing atmospheric conditions or by up-wind sources, an average background is determined based on reference measurements performed upwind the landfill before and after the measuring campaign (before and after plume traverses). In the case, where the background concentrations are varying over time and space, the background is assessed for each individual plume using the background concentration on each side of the plume. A linear extrapolation of the background concentration is thus made – meaning that the background concentration is assumed to increase/decrease linearly from plume start until plume end.

The impact of the linear assumption depends highly on the change in background concentration and the emission/dilution from the landfill. We have previous looked closer at this and found that the influence was limited (< 5%) when the plume was more than 10 times higher than the change in background concentration.

To evaluate the influence of different ways to determine and subtract the background concentration of both methane and tracer gas on the emission results obtained in this study, the dataset from the measuring campaign performed on the morning of November 13th (landfill M) was processed in two different ways:

1) Subtracting a constant average background concentration from all plume concentrations. The average background concentration was calculated as the average of the concentration in the air before entering the first plume in the measurement series and the concentration measured after the last plume of the series. Care was taken that no interfering sources influenced the concentration of methane and tracer gas during the background measurements.
2) Subtracting an average background concentration from each individual plume. The average background concentration was calculated as the average of the concentration in the air before entering and after leaving each plume. This approach, evaluating each plume to find the background concentration in the given situation, is considered the most accurate and is the one that was used to when calculating the emissions in this study (shown earlier in the report).

Table 15 shows an overview of the calculated emission rates using the two different approaches for determining the background concentration of methane and acetylene. Using the first approach

calculation one constant background for methane gave a background concentration of 1938 ppb (average of before first plume (1934 ppb) and after last plume (1942 ppb)). Similar, one constant background for acetylene gave a background concentration of 0.39 ppb (average of before first plume (0.53 ppb) and after last plume (0.26 ppb)). The background concentration of the tracer gas was thus higher before the series than after and the background concentration of methane was higher after the series than before. For the second approach the background concentrations varied for each individual plume.

Table 15 shows the influence on the calculated methane emission rate when using two different approaches for determining the background concentrations of methane and acetylene. It can be seen that the average emission for the two situations are very different; the first approach giving a smaller emission (7.9 kg h^{-1}) than the second approach (13.5 kg h^{-1}). The reason for the difference is mainly an increase in the methane background concentration during the last part of the measurement series causing a too high background subtraction in the first 2/3 of the measurements, while a too small background concentration in the last third of the measurement. At the same time, there is a decreasing tracer gas background concentration giving the same effect on the calculated methane emission rate.

Table 15. The influence on the methane emission rate when using two different approaches for determining the background concentrations of methane and acetylene. Note that the individual background approach is the one taken in the report.

Time	Measured methane emission rate (kg h^{-1})	
	One constant average background for the whole time series (all plumes)	Individual backgrounds for each plume traverse
11.07	6.1	11.2
11.11	6.3	13.6
11.14	3.9	17.1
11.16	2.7	11.7
11.18	6.2	15.6
11.20	6.5	14.8
11.22	6.8	14.6
11.25	10.0	12.4
11.27	13.9	12.7
11.29	16.4	10.8
Average	**7.9**	**13.5**
Std deviation	**4.3**	**2.1**
Std error of mean	**1.4**	**0.7**
Degree of freedom	**9**	**9**
95% confidence interval	**±3.1**	**±1.5**

Influence of wind direction and wind speed

At landfill M, emissions were measured on 11th, 12th and 13th of November. The highest emissions were measured on the 11th and a difference in emission between morning and afternoon was observed on 13th. The DIAL team from NPL recorded wind data (speed and direction) during the three day measuring campaign but only when they performed their own measurements. On the 11th, the DIAL team stopped measuring around 16.30 and the tracer team measured their first useful traverse at 18.00. There is therefore no wind data available for the emission rates measured by the tracer team on the 11th. Wind data are available for all plume traverses performed on the 12th. On the 13th, the DIAL team measured the wind from 8.30 to

16.00, while the tracer team measured emissions from 11.00 to 11.30 and from 17.00 to 18.15. There is therefore only wind data available for the measurements performed early in the day on the 13th.

Table 16. Overview of available weather data provided by the DIAL team.

Day	Time intervals for available weather data from the DIAL team available	Time intervals for measurements performed by the tracer team	Weather data available (yes/no)
11th, November	09:08 – 16:32	18:00 – 19:15	no
12th , November	09:07 – 16:22	12:30 – 14.30	yes
13th , November	08:31 – 16:08	11:00 – 11:30 16:45 – 18:15	yes no

The two measurement periods with wind data have different average emission rates: 6.6±0.5 kg h^{-1} on the 12th and 13.5±1.5 kg h^{-1} on the 13th before noon. On both days, there were smaller variations in the measured emissions within the measurement series, but no trend throughout the series. On the 12th, the average wind speed for the measurement period was measured to 6.9 m/s by the DIAL team at the top of their mast, while a nearby private weather station measured an average wind speed of 4.2 m s^{-1}. Based on these two wind speeds is was estimated that the measured plumes 1500-2000 m downwind from the landfill was emitted between 3.6 and 8.0 minutes before they were measured. The DIAL wind measurements have a temporal resolution of 1 min and varied several m/s from one minute average to another. It is not possible to capture this variation in the emission rates for the following reasons: 1) a plume traverse takes several minutes to perform and the calculated emission is therefore the average emission in those minutes, 2) it is not possible to assign a specific emission timestamp to the measured plumes, as the wind speed and measurement distance vary during the measurement. When a plume is measured and assigned a timestamp, this time is the time when the centre of the plume is measured. A plume often takes 1-3 minutes to measure, which makes the calculated emission rate an average of the emission during those minutes. Additionally, the uncertainty in plume transport time (3.6 to 8.0 minutes) makes the actual emission time more uncertain.

As there is no significant trend in the wind speed or directions during the two days with matching weather and emission data, no valuable information on the emission's dependence on weather changes can be derived from the measurements at landfill M.

The DIAL group did not measure the atmospheric pressure. A comparison with the pressure measured at a local weather station is shown elsewhere in the report. As the measurements were performed with and without flaring of the landfill gas and the effect of this flaring on the total emission is unknown, it can be difficult to assign the emission variation to a specific variable.

The emission at landfill J was measured by the tracer team on November 17th, 18th, 19th and 20th. The DIAL team measured weather data on 18th to 20th. Figure 28 shows scatter plots of the individual emission rates and the corresponding weather data. For the reasons explained earlier in the text about landfill M, it is not possible to assign a specific emission time to a measured transect, but in the following graphs the average wind speed was used to estimate the "delay" between emission and measurement time. As some of these measurements were performed more than 5 km from the landfill and the wind speed was low, time delay of up to 45 min were considered.

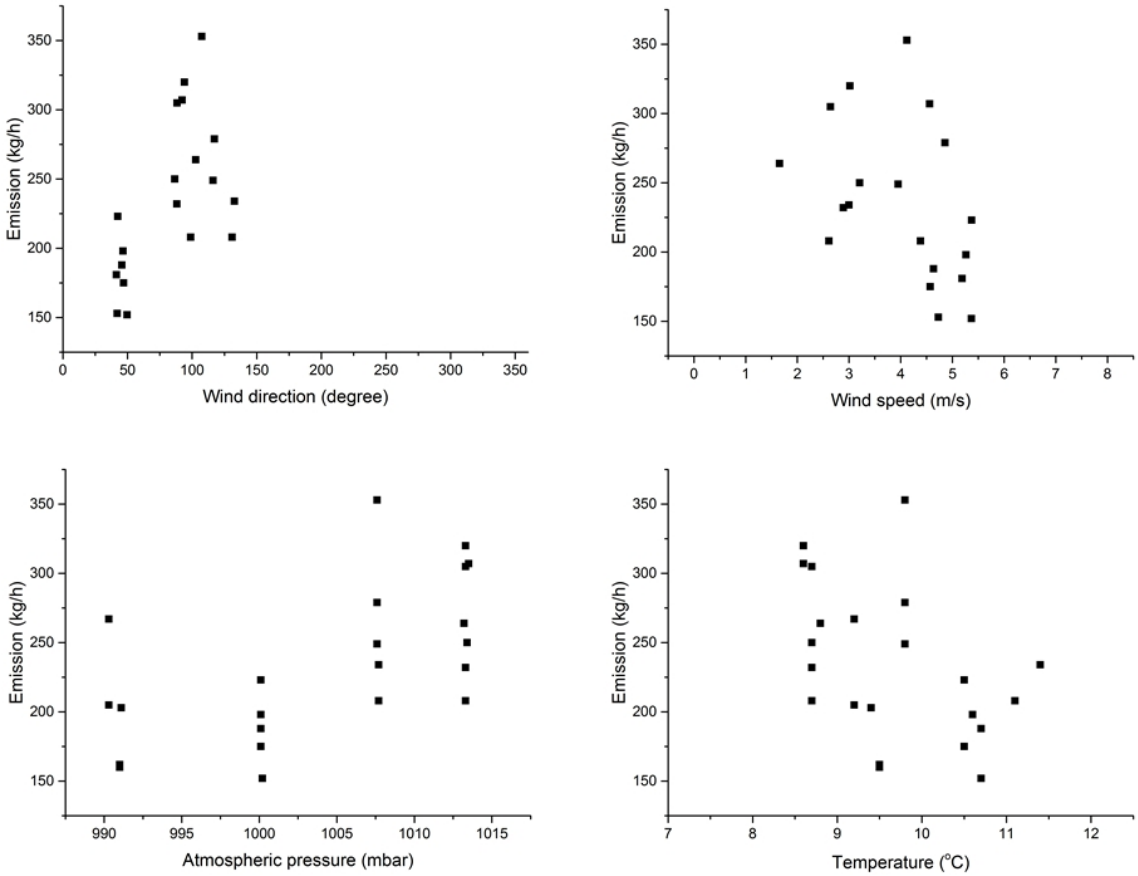

Figure 28. Correlation plots between measured methane emission rates (kg h^{-1}) and weather conditions (wind direction, wind speed, atmospheric pressure, and temperature.

The emission variation within the measurement series/days could not clearly be explained by the atmospheric conditions and changes of these. Lower emission was measured at a wind direction around 50 degrees than when the wind came from 90 to 135 degrees, so it cannot be ruled out that the wind direction have an influence on either the emission rate or the measurement performance, but the data in this measurement period is insufficient to make any conclusions.

The possible influence of barometric pressure on the measured landfill emission rates are presented and discussed in the individual sections presenting the results. The conclusion that the emission can be influenced by the atmospheric pressure is not based on the actual measurements, but on the literature and previous measurements using the tracer dispersion method. There is limited knowledge on the emission response time to a change in barometric pressure, wind speed or change in gas extraction rate. The response time to such changes is most likely site specific. However, there are many clear cases showing that pressure changes can have a significant influence on the methane emission and that this can happen relatively fast (again, depending on the landfill and the emission routes (cracks, wells, biocovers, gas extraction, etc.)). Most investigations on emissions change due to pressure change are based on surface flux measurements. The big study measuring whole site emission response to pressure changes or operation changes is simply lacking.

Source simulations, tracer coverage, number of traverses and uncertainty
The variation seen between individual transects measured in the same series is the sum of the overall uncertainty and the actual emission variation, and is difficult to quantify as it will depend on the actual landfill and the conditions present on the measurement day and in general. For some of the measuring campaigns, the variation between individual transects can seem large, but with multiple transects, this uncertainty becomes significantly lower (when the variation is random and not an emission trend). For this reason, we always recommend to perform a minimum of 10 successful transects for each quantification (e.g. compared to the DiAL team's three flux measurements from each area). The statistics based on more than 10 transects gives an uncertainty of the measured emission rate, which is significantly lower than the estimated overall uncertainty. For comparison it can be noted that also the DiAL occasionally has similar variation between individual flux measurements. Unfortunately, the conditions during the measurement campaigns did not allow enough successful transects on the individual measurement days. At this study it was not possible to choose the specific measuring day or the measuring time of day needed. A less sunny day with more (and stable) wind would have allowed many more transects and thus a significantly better quantification. Obviously having a wider time frame for measurement performance would allow better planning and consideration of the weather conditions (including taking advantage of the changing wind directions) for optimal quantifications at the individual landfills.

The number of tracer gas bottles was small at landfill J and thus the methane emission was not well covered by the tracer gas release. This was stressed already from the beginning of the measurement, but the amount of measurement days, the physical size of the landfill, the limited accessibility to the different landfill areas and the limited number of tracer gas bottles (including that two of the delivered bottles were broken) made it a difficult talk to make optimal measurements. The number of tracer gas bottles that should be used depends mainly on the nature of the methane emission (number of emission areas, and the magnitude of these emissions) and less on the size of the landfill, although the size also play a role, since bigger sizes require larger distances to the measurement road to consider the landfill as a point source.

8. Further recommendations

Observations from UK measurements
The tracer dispersion method has been applied at more than 20 Danish landfills for methane quantifications. The UK landfills are in general larger both in terms of area, disposal height, disposed waste amounts including higher amounts of organic waste and thus also in terms of methane emissions. During 2014, the tracer dispersion method has been applied at four UK landfills for quantification of whole site methane emissions. One of the purposes was to investigate the application of the tracer dispersion method at larger landfill sites. The measuring campaigns were performed within a few days - at one site only one day of measurements was conducted. No initial information about landfill layout, emission areas, on-site sources etc. were provided before the measuring campaigns. At a relatively small and closed landfill, a good tracer-methane correlation was obtained. Methane screening measurements often revealed small surrounding sources such as piles of manure, leaks from gas pipes and emission from various farm houses. At one site, methane screening measurements also revealed a larger upwind methane source approximately 5 km from the landfill, which interfered with the measurements on a single measurement day when the wind came directly from there. However, it was possible to

subtract the additional methane source from the downwind plume and estimate the total methane emission from the landfill. It is however important to be aware of other local methane sources, which can interfere with the landfill methane plume. This is also a potential problem with other remote measurement methods. Again with a broader time period for measurement performance, measurements could be conducted using more optimal wind directions avoiding the influence from surrounding sources.

Uncertainty sources

The tracer dispersion method is sensitive to the simulation of the source – accurate placement of the tracer release bottles. The tracer gas bottles should be placed as close to the main methane sources at the landfill as possible. Misplacing the tracer gas bottles will increase the uncertainty of the measurement, although this uncertainty will decrease with increasing plume dispersion, thus at more unstable weather conditions and/or when measurements are conducted further away from the landfill.

A part of the standard procedure is to locate the main emission areas by performing an onsite methane screenings. Most of larger landfills studied so far in the UK have limited access for mobile on-site measurements due to lack of suitable roads on the landfill, safety restrictions, or due to ongoing activities at the landfill. If pervious landfill walkover surveys have been performed, information from these surveys about emission hotspots could be used for a more accurate tracer gas placement. Also knowledge about leaking areas should be provided for better tracer gas placement.

To test how well the tracer gas bottles have been placed, traverses of the downwind plume relatively close to the landfill can be done, revealing if the tracer gas bottles are misplaced lateral compared to the main emission area. The same test can be done with a different wind direction to see if the tracer gas bottle is misplaced in the opposite direction.

It is suggested that a significant effort is put into placing the tracer gas bottles in the right way. Furthermore, it is suggested that future studies will focus on the effect of misplacing the tracer gas bottles at large landfills with multiple emission areas. It is also suggested to evaluate the number of tracer gas bottles needed, depending on the landfill size, different sections at the landfill and the methane emission from these sections.

Temporal variation in landfill emissions

It is well-established that landfill emissions are influenced by changes in the meteorological conditions especially atmospheric pressure changes and wind speeds (Czepiel et al., 2003; Gebert & Groengroeft 2006; Rachor et al., 2013; Poulsen et al, 2001; Poulsen & Møldrup, 2006). However, there is still a lack of knowledge about the dynamics in landfill emissions – magnitude and frequency of emission response due to pressure changes (as an example). It is advised that future studies focus on evaluation the temporal emission variation from whole site as well as from different landfills sections, as these may vary significantly depending on weather conditions such as precipitation, atmospheric pressure, wind and temperature. Additionally, information about how methane emission from a landfill changes with changes in landfill gas extraction would be very useful in order to evaluate when and how a good representative measurement of the methane emission from the landfill can be performed.

9. References

Börjesson, G., Samuelsson, J., Chanton, J. (2007). Methane oxidation in Swedish landfills quantified with the stable carbon isotope technique in combination with an optical method for emitted methane. *Environmental Science & Technology*, *41*, 6684–6690.

Börjesson, G., Samuelsson, J., Chanton, J., Adolfsson, R., Galle, B., Svensson, B.H. (2009). A national landfill methane budget for Sweden based on field measurements, and an evaluation of IPCC models. *Tellus B*, *61*, 424–435.

Czepiel, P.M., Shorter, J.H., Mosher, B., Allwine, E., McManus, J.B., Harriss, R.C., … Lamb, B.K. (2003). The influence of atmospheric pressure on landfill methane emissions. *Waste Management*, 23 (7), 593–598.

Foster-Wittig, T.A., Thoma, E.D., Green, R.B., Hater, G.R., Swan, N.D., Chanton, J.P. (2015). Development of a mobile tracer correlation method for assessment of air emissions from landfills and other area sources. *Atmospheric Environment*, 102, 323–330.

Galle, B., Samuelsson, J., Svensson, B.H., Börjesson, G. (2001). Measurements of methane emissions from landfills using a time correlation tracer method based on FTIR absorption spectroscopy. *Environmental Science & Technology* 35 (1), 21-25.

Gebert, J., Groengroeft, A. (2006). Passive landfill gas emission - Influence of atmospheric pressure and implications for the operation of methane-oxidising biofilters. *Waste Management*, 26 (3), 245–251.

Mønster, J.G., Samuelsson, J., Kjeldsen, P., Rella, C.W. Scheutz, C. (2014). Quantifying methane emission from fugitive sources by combining tracer release and downwind measurements – a sensitivity analysis based on multiple field surveys. *Waste Management*, 34 (8), 1416-1428.

Mønster, J., Samuelsson J., Kjeldsen, P., Scheutz, C. (2015). Quantification of methane emissions from 15 Danish landfills using the mobile tracer dispersion method. *Waste Management*, 35, 177-186.

Poulsen, T.G., Christophersen, M., Moldrup, P., Kjeldsen, P. (2001). Modeling lateral gas transport in soil adjacent to old landfill. *J. Environ. Eng*. 127, 145-153.

Poulsen, T.G., Møldrup, P. (2006). Evaluating effects of wind-induced pressure fluctuations on soil–atmosphere gas exchange at a landfill using stochastic modelling. *Waste Management and Research*, 24, 473–481.

Rachor, I.M., Gebert, J., Gröngröft, A., Pfeiffer, E.-M. (2013). Variability of methane emissions from an old landfill over different time-scales. *European Journal of Soil Science*, 64(1), 16–26.

Roscioli, J.R., Yacovitch, T.I., Floerchinger, C., Mitchell, A.L., Tkacik, D.S., Subramanian, R., … Marchese, A.J. (2014). Measurements of methane emissions from natural gas gathering

facilities and processing plants: measurement methods. *Atmospheric Measurement Techniques Discussions*, 7 (12), 12357–12406.

Scheutz, C., Samuelsson, J., Fredenslund, A. M., and Kjeldsen, P. (2011). Quantification of multiple methane emission sources at landfills using a double tracer technique. *Waste Management*, 31 (5), 1009-1017.